

Considerations on the quantum double-exchange Hamiltonian

A. Weiße,¹ J. Loos,² and H. Fehske¹

¹*Physikalisches Institut, Universität Bayreuth, 95440 Bayreuth, Germany*

²*Institute of Physics, Czech Academy of Sciences, 16200 Prague, Czech Republic*

(Received 16 January 2001; published 2 July 2001)

Schwinger bosons allow for an advantageous representation of quantum double exchange. We review this subject, comment on previous results, and address the transition to the semiclassical limit. We derive an effective fermionic Hamiltonian for the spin-dependent hopping of holes interacting with a background of local spins, which is used in a related publication within a two-phase description of colossal magnetoresistant manganites.

DOI: 10.1103/PhysRevB.64.054406

PACS number(s): 75.10.-b, 75.30.Et, 75.30.Vn

I. INTRODUCTION

Introduced by Zener¹ in the early 1950s, the notion of double exchange together with mixed-valency manganites $R_{1-x}A_x\text{MnO}_3$ (where $R=\text{La, Pr, Nd}$ and $A=\text{Sr, Ca, Ba, Pb}$) attracted renewed attention when a colossal magnetoresistive effect was discovered in these compounds some years ago.² The magnetic and electronic properties of manganese oxides, to some extent, are believed to arise from the large Coulomb and Hund's rule interaction of the manganese d shell electrons. Due to the almost octahedral coordination within the perovskite structure the d levels split into two subbands labeled according to their octahedral symmetry, e_g and t_{2g} . In the case of zero doping ($x=0$), there are four electrons per Mn site that fill up the three t_{2g} levels and one e_g level, and by Hund's rule, form a $S=2$ spin state. Doping will remove the electron from the e_g level, and by hopping via bridging oxygen sites these holes acquire mobility. However, this hopping acts in a background of local spins $S=3/2$ formed by the t_{2g} electrons and its amplitude depends on the overlap of the spin states at neighboring sites (or, in a classical language, on their relative angle); it is largest if the total bond spin is maximal and vice versa.³

Another ingredient, which is assumed to significantly influence the physical properties of manganites, is electron-lattice interaction. Namely, the two e_g orbitals, which are degenerate in a perfect cubic environment, will couple to lattice vibrations of the same symmetry, giving rise to a Jahn-Teller effect and polaronic behavior in some regions of the phase diagram. It is this close interplay of three different subsystems (electrons in degenerate orbitals, background of localized spins, and lattice vibrations) that makes the physics of manganites both rich and complicated.

In the present work we concentrate on the double exchange (DE) part of the interactions and consider different possibilities for an approximate treatment of the exact DE Hamiltonian on a lattice in terms of effective electronic one- or two-band models. These can be used in a more elaborate modeling of the real materials (see our forthcoming work⁴). It turns out that quantum double exchange on a lattice is most suitably derived and described with the help of Schwinger bosons. We therefore include a detailed and pedagogic derivation of the quantum DE Hamiltonian using Schwinger bosons. Although this approach has been used

before,⁵ we feel a comprehensive presentation of the subject is still missing. In two appendixes we reexamine the derivation for two sites, and consider the semiclassical limit ($S\rightarrow\infty$). In addition, by means of numerical experiments, we illustrate how this limit evolves from the quantum case.

II. SCHWINGER BOSON REPRESENTATION OF DOUBLE EXCHANGE

To derive the quantum DE Hamiltonian on a lattice, as a starting point we take the Kondo lattice model including on-site Coulomb repulsion,

$$H = -t \sum_{\langle ij \rangle \sigma} [c_{i\sigma}^\dagger c_{j\sigma} + \text{H.c.}] - J_H \sum_{i\sigma\sigma'} (\mathbf{S}_i \cdot \boldsymbol{\sigma}_{\sigma\sigma'}) c_{i\sigma}^\dagger c_{i\sigma'} + U \sum_i n_{i\downarrow} n_{i\uparrow}, \quad (1)$$

where summation is over nearest-neighbor bonds $\langle ij \rangle$ or sites i , respectively. For clarity and since it can be included easily in the final result, here we have neglected the orbital degeneracy of the e_g electrons. That is, $c_{i\sigma}^{(\dagger)}$ denote electrons in a single band, which interact with some localized spin \mathbf{S}_i via the Hund's coupling J_H . In the real materials this localized spin corresponds to the remaining t_{2g} electrons, which tend to form a high spin state with an electron in the e_g shell.

In the manganites the situation is such that $U \gg J_H > t$ (cf. Refs. 3 and 6). Hence, we first take the limit $U \rightarrow \infty$, resulting in

$$H = -t \sum_{\langle ij \rangle \sigma} [\tilde{c}_{i\sigma}^\dagger \tilde{c}_{j\sigma} + \text{H.c.}] - J_H \sum_{i\sigma\sigma'} (\mathbf{S}_i \cdot \boldsymbol{\sigma}_{\sigma\sigma'}) \tilde{c}_{i\sigma}^\dagger \tilde{c}_{i\sigma'} \quad (2)$$

with restricted fermions $\tilde{c}_{i\sigma} = c_{i\sigma}(1 - n_{i,-\sigma})$. Next, following Kubo and Ohata,⁷ the exchange term in Eq. (2) is solved while the hopping term is considered as a small perturbation. For positive J_H the ground state of the exchange term is a free spin S , if there is no electron at site i , or a coupled spin $\bar{S} = S + 1/2$ otherwise (note that we use S for the length of the localized spin \mathbf{S}_i formed by t_{2g} electrons). To describe the effective hopping we therefore need a projection operator, which restores these conditions,

$$(P_i^+)_{\sigma\sigma'} = \frac{(\mathbf{S}_i \cdot \boldsymbol{\sigma}_{\sigma\sigma'}) + (S+1)\delta_{\sigma\sigma'}}{2S+1}. \quad (3)$$

Then the DE Hamiltonian [Eq. (2.3) in Ref. 7] is given in terms of spin and restricted fermion operators by

$$H_{\text{el}}^{\text{DE}} = -t \sum_{\langle ij \rangle \sigma \sigma'} [\tilde{c}_{i\sigma}^\dagger (P_i^+ P_j^+)_{\sigma\sigma'} \tilde{c}_{j\sigma'} + \text{H.c.}]. \quad (4)$$

However, this expression turns out to be unwieldy for analytic as well as numeric calculations. Although in principle the electronic spin is absorbed into the total spin at each site, the spin index σ is still present in Eq. (4). Here the advantages of Schwinger bosons come into play, namely, the possibility to describe spins of arbitrary amplitude with the same set of boson operators a_i and b_i ,

$$S_i^+ = a_i^\dagger b_i, \quad S_i^- = b_i^\dagger a_i, \quad (5)$$

$$S_i^z = (a_i^\dagger a_i - b_i^\dagger b_i)/2, \quad (6)$$

$$|S_i| = (a_i^\dagger a_i + b_i^\dagger b_i)/2. \quad (7)$$

Using these operators, we can rewrite the projection operators P_i^+ ,

$$\begin{aligned} (P_i^+)_{\sigma\sigma'} &= \frac{1}{2S+1} \begin{bmatrix} (S+1) + S_i^z & S_i^- \\ S_i^+ & (S+1) - S_i^z \end{bmatrix} \\ &= \frac{1}{2S+1} \begin{bmatrix} a_i a_i^\dagger & a_i b_i^\dagger \\ b_i a_i^\dagger & b_i b_i^\dagger \end{bmatrix}, \end{aligned} \quad (8)$$

where we can keep S in the denominator, because it is conserved. The last matrix can be decomposed easily,

$$(P_i^+)_{\sigma\sigma'} = \frac{1}{2S+1} \begin{bmatrix} a_i \\ b_i \end{bmatrix} \cdot [a_i^\dagger \quad b_i^\dagger], \quad (9)$$

which leads to

$$H_{\text{el}}^{\text{DE}} = \frac{-t}{2S+1} \sum_{\langle ij \rangle} [(R_i^+)^\dagger (a_i^\dagger a_j + b_i^\dagger b_j) R_j^+ + \text{H.c.}] \quad (10)$$

with the projectors

$$R_i^+ = \frac{\tilde{c}_{i\uparrow} a_i^\dagger + \tilde{c}_{i\downarrow} b_i^\dagger}{\sqrt{2S+1}}. \quad (11)$$

If we restrict the Hilbert space of the problem by fixing the spin length at each site to the value $S + n_i/2$ (where $n_i = \tilde{n}_{i\uparrow} + \tilde{n}_{i\downarrow}$), we can simply replace the projectors R_i^+ by spinless fermion operators c_i . This becomes clear by taking a closer look on the operators R_i^+ . Let us assume that site i is occupied by an electron, and therefore the total spin at the site is $S + 1/2$. The corresponding state $|n_e; S, m\rangle$ can be written as

$$\begin{aligned} |1; S + \frac{1}{2}, m\rangle &= \sqrt{\frac{S + \frac{1}{2} + m}{2S+1}} |\uparrow\rangle |S, m - \frac{1}{2}\rangle \\ &+ \sqrt{\frac{S + \frac{1}{2} - m}{2S+1}} |\downarrow\rangle |S, m + \frac{1}{2}\rangle. \end{aligned} \quad (12)$$

Using the representation of an S^z eigenstate in terms of Schwinger bosons,

$$|S, m\rangle = \frac{(a^\dagger)^{S+m} (b^\dagger)^{S-m}}{\sqrt{(S+m)!(S-m)!}} |0\rangle, \quad (13)$$

we find that by applying the operator R^+ , the state $|1; S + 1/2, m\rangle$ in Eq. (12) is transformed into the corresponding Schwinger boson representation of the coupled spin $S + 1/2$, while the electron is annihilated,

$$R^+ |1; S + \frac{1}{2}, m\rangle = |0; S + \frac{1}{2}, m\rangle. \quad (14)$$

Backwards, the operator $(R^+)^\dagger$ creates the decomposition of the coupled spin into electronic and localized spin, i.e., it produces appropriate Clebsch-Gordan coefficients.

Consequently, if the electronic spin merges into the Schwinger boson representation via the constraint, we can omit the index σ of the fermions. What remains of the operators R_i^+ are spinless fermions, c_i , which automatically satisfy the restriction on the local electron number. The corresponding Hamiltonian is given by

$$H_{\text{el}}^{\text{DE}} = \frac{-t}{2S+1} \sum_{\langle ij \rangle} [(a_i^\dagger a_j + b_i^\dagger b_j) c_i^\dagger c_j + \text{H.c.}], \quad (15)$$

where, for every site i , the Hilbert space is constrained to

$$a_i^\dagger a_i + b_i^\dagger b_i = 2S + c_i^\dagger c_i. \quad (16)$$

In the case of low doping usually it is more appropriate and natural to consider holes instead of electrons. Here ‘‘hole’’ denotes a fermion and a spin S moving together in a background of spins $\bar{S} = S + 1/2$. For the transformation of the electronic Hamiltonian, Eq. (10), the operator R^+ needs to be replaced by a suitable counterpart involving restricted hole operators $\tilde{h}_{i\sigma}$. In analogy to Eqs. (12)–(14), the state

$$\begin{aligned} |n_h = 1; S, m\rangle &= \sqrt{\frac{\bar{S} - m + \frac{1}{2}}{2\bar{S}+1}} |\uparrow\rangle |\bar{S}, m - \frac{1}{2}\rangle \\ &- \sqrt{\frac{\bar{S} + m + \frac{1}{2}}{2\bar{S}+1}} |\downarrow\rangle |\bar{S}, m + \frac{1}{2}\rangle \end{aligned} \quad (17)$$

is transformed into its corresponding Schwinger boson representation by the operator

$$R_i^- = \frac{\tilde{h}_{i\uparrow} b_i - \tilde{h}_{i\downarrow} a_i}{\sqrt{2\bar{S}+1}}. \quad (18)$$

Hence, in hole representation, the DE Hamiltonian reads

$$H_{\text{hole}}^{\text{DE}} = \frac{t}{2\bar{S}} \sum_{\langle ij \rangle} [(R_i^-)^\dagger (a_i a_j^\dagger + b_i b_j^\dagger) R_j^- + \text{H.c.}]. \quad (19)$$

The Hamiltonian for spinless fermions, Eq. (15), changes only little, becoming

$$H_{\text{hole}}^{\text{DE}} = \frac{t}{2\bar{S}} \sum_{\langle ij \rangle} [(a_i a_j^\dagger + b_i b_j^\dagger) h_i^\dagger h_j + \text{H.c.}] \quad (20)$$

with the constraint

$$a_i^\dagger a_i + b_i^\dagger b_i = 2\bar{S} - h_i^\dagger h_i. \quad (21)$$

III. EFFECTIVE TRANSPORT HAMILTONIAN

To obtain an effective Hamiltonian for the spin-dependent hole-hopping, the spin part of the DE interaction is considered within mean-field approximation. However, there are two representations of the DE Hamiltonian to start from: Eqs. (19) and (20). The resulting effective Hamiltonians describe carriers with or without spin, respectively. Given Eq. (20), a mean hopping of the spinless carriers, $\tilde{t}^{(b)}$, is obtained by considering each bond $\langle ij \rangle$ separately, and taking the expectation value of the DE term for all values of the total bond spin S_T and S_T^z in an effective ordering field $\lambda = \beta g \mu_B H_{\text{eff}}^z$. This reproduces the result of Kubo and Ohata.⁷ Having spin \bar{S} at site i and spin S at site j , we may consider the coupled state S_T of maximal S_T^z , given by

$$|S_T, S_T\rangle_{\langle \bar{S} S \rangle} = C (b_i^\dagger a_j^\dagger - b_j^\dagger a_i^\dagger)^{2S+1/2-S_T} (a_i^\dagger)^{S_T+1/2} \times (a_j^\dagger)^{S_T-1/2} |0\rangle, \quad (22)$$

with the normalization factor

$$C = \frac{[(2S_T+1)!]^{1/2}}{\left(S_T + \frac{1}{2}\right)! \left(S_T - \frac{1}{2}\right)! \left(2S + \frac{1}{2} - S_T\right)! \left(2S + \frac{3}{2} + S_T\right)!} \quad (23)$$

Applying the DE expression $(a_j^\dagger a_i + b_j^\dagger b_i)$, which commutes with $S_T^+ = a_i^\dagger b_i + a_j^\dagger b_j$ and S_T^- , we arrive at

$$\begin{aligned} & (a_j^\dagger a_i + b_j^\dagger b_i) |S_T, S_T\rangle_{\langle \bar{S} S \rangle} \\ &= C \left(S_T + \frac{1}{2}\right) (b_i^\dagger a_j^\dagger - b_j^\dagger a_i^\dagger)^{2S+1/2-S_T} \\ & \quad \times (a_i^\dagger)^{S_T-1/2} (a_j^\dagger)^{S_T+1/2} |0\rangle \\ &= \left(S_T + \frac{1}{2}\right) |S_T, S_T\rangle_{\langle \bar{S} S \rangle}. \end{aligned} \quad (24)$$

Hence, we rederived the S_T^z -independent effective matrix element for a single bond (cf. Appendix A or Ref. 3),

$$t^{(b)} = \frac{S_T+1/2}{2\bar{S}} t, \quad (25)$$

which is averaged over all values and directions of S_T ,

$$\tilde{t}^{(b)} = \gamma_{\bar{S}}[\bar{S}\lambda] t, \quad (26)$$

where⁷

$$\begin{aligned} \gamma_{\bar{S}}[\bar{S}\lambda] &= \frac{\sum_{S_T=1/2}^{2\bar{S}-1/2} \sum_{M=-S_T}^{S_T} \frac{S_T+1/2}{2\bar{S}} e^{M\lambda}}{\sum_{S_T=1/2}^{2\bar{S}-1/2} \sum_{M=-S_T}^{S_T} e^{M\lambda}} \\ &= \frac{1}{2} + \frac{\bar{S}}{2\bar{S}+1} \coth\left(\frac{2\bar{S}+1}{2}\lambda\right) \\ & \quad \times \left[\coth(\bar{S}\lambda) - \frac{1}{2\bar{S}} \coth\left(\frac{\lambda}{2}\right) \right]. \end{aligned} \quad (27)$$

The effective Hamiltonian describing spinless fermionic holes in an averaged background of ordered spins reads

$$H_{\text{hole}}^{\text{eff, I}} = \tilde{t}^{(b)} \sum_{\langle ij \rangle} [h_i^\dagger h_j + \text{H.c.}]. \quad (28)$$

In Appendix B we compare the classical limit of both this Hamiltonian and the exact expression. At least the bandwidth turns out to be represented very well.

Another possible way to obtain an effective Hamiltonian is based on the picture of itinerant carriers of spin $\frac{1}{2}$ moving in the background of localized spins, the correlations of which change on a large time scale compared with the hopping frequency. Then an effective hopping Hamiltonian is obtained averaging $H_{\text{hole}}^{\text{DE}}$, Eq. (19), over free spins \bar{S} in a homogeneous field λ . According to Eqs. (5) and (6) and the fact that $\langle S^\pm \rangle = 0$ and $\langle S^z \rangle = \bar{S} B_{\bar{S}}[\bar{S}\lambda]$ (where $B_{\bar{S}}[z]$ denotes the Brillouin function), only two terms contribute. The resulting Hamiltonian involves two effective hopping matrix elements, one for each spin channel,

$$H_{\text{hole}}^{\text{eff, II}} = \sum_{\langle ij \rangle} [\tilde{t}_\uparrow \tilde{h}_\uparrow^\dagger \tilde{h}_\uparrow + \tilde{t}_\downarrow \tilde{h}_\downarrow^\dagger \tilde{h}_\downarrow + \text{H.c.}] \quad (29)$$

with

$$\tilde{t}_\uparrow = \frac{t}{(2\bar{S})(2\bar{S}+1)} \{\bar{S}(1 - B_{\bar{S}}[\bar{S}\lambda])\}^2, \quad (30)$$

$$\tilde{t}_\downarrow = \frac{t}{(2\bar{S})(2\bar{S}+1)} \{\bar{S}(1 + B_{\bar{S}}[\bar{S}\lambda])\}^2. \quad (31)$$

In a fully polarized background ($\lambda \rightarrow \infty$) only holes with anti-parallel spin can hop ($\tilde{t}_\downarrow \rightarrow [2\bar{S}/(2\bar{S}+1)]t$), while holes with parallel spin are blocked ($\tilde{t}_\uparrow \rightarrow 0$). In general, the situation is complicated by the fact that Eq. (29) involves restricted fermion operators (Hubbard operators) preventing an exact solution of the model. However, to a good approximation, in the polarized phase the spin-up band can be neglected, while the spin-down band is taken into account using unrestricted operators $h_{i\downarrow}^{(\dagger)}$. On the other hand, in a

disordered phase ($\lambda \rightarrow 0$) both bands are equivalent ($\tilde{t}_\uparrow = \tilde{t}_\downarrow \rightarrow [\bar{S}/(4\bar{S}+2)]t$), making an approximate treatment less evident.

IV. CONCLUSIONS

In the present work we review the subject of double exchange and derive an effective Hamiltonian for the spin-dependent hopping of holes in an averaged background of local spins. This is used in a subsequent publication within a two-phase scenario for the description of colossal magnetoresistant manganites,⁴ alternatively to the effective Hamiltonian for spinless carriers derived in Ref. 7. Besides, we illustrate that on the level of quantum spins all features of double exchange—namely, its derivation, limiting cases, as well as approximations—are probably most clearly represented in terms of Schwinger bosons.

ACKNOWLEDGMENTS

This work was supported by the Deutsche Forschungsgemeinschaft and the Czech Academy of Sciences under Grant No. 436 TSE 113/33.

APPENDIX A: REEXAMINATION OF THE TWO-SITE PROBLEM

As noted above, the DE matrix element, Eq. (25), was first derived by Anderson and Hasegawa³ by considering a system of two spins S at neighboring sites and a mobile electron whose spin σ is coupled to the local spin at the same site. Recently the problem was reexamined by Müller-Hartmann and Dagotto,⁸ who found a “nontrivial” phase factor, which the authors interpreted in terms of a Berry phase within the limit $S \rightarrow \infty$. We argue that the phase factor of their quantum DE Hamiltonian merely compensates for the phase introduced by the permutation of neighboring spin states, whereas independently the Berry phase evolves in the classical ($S \rightarrow \infty$) limit of the correct DE Hamiltonian, Eq. (15). Moreover, on a lattice, double exchange cannot be described in terms of spin and permutation operators, which do not take into account fermionic commutation relations.

To be specific we briefly summarize the two-site problem. Assuming the electron initially to be at site 1 we start with coupling the local spin \mathbf{S}_1 and the electron spin $\boldsymbol{\sigma}$, to give an on-site spin $\bar{\mathbf{S}}_1 = \mathbf{S}_1 + \boldsymbol{\sigma}$. We note that the construction of the corresponding wave function is not unique, as is known from textbooks on spin algebra.⁹ In more detail,

$$|\bar{S}_1 \bar{m}_1\rangle_{(\sigma S_1)} = \sum_{\mu m_1} \begin{pmatrix} \sigma & S_1 & \bar{S}_1 \\ \mu & m_1 & \bar{m}_1 \end{pmatrix} |\sigma \mu\rangle |S_1 m_1\rangle, \quad (\text{A1})$$

$$|\bar{S}_1 \bar{m}_1\rangle_{(S_1 \sigma)} = (-1)^{\sigma + S_1 - \bar{S}_1} |\bar{S}_1 \bar{m}_1\rangle_{(\sigma S_1)}, \quad (\text{A2})$$

where we give the order of the constituting spin states as a subscript and denote Clebsch-Gordan coefficients by $(\cdot \cdot | \cdot)$. Adding this spin to the unchanged core spin at site 2, we arrive at an initial state of total spin S_T

$$|\varphi_{\text{in}}\rangle = |S_T m_T\rangle_{((\sigma S_1) S_2)} \quad (\text{A3})$$

$$= \sum_{\bar{m}_1 m_2} \begin{pmatrix} \bar{S}_1 & S_2 & S_T \\ \bar{m}_1 & m_2 & m_T \end{pmatrix} |\bar{S}_1 \bar{m}_1\rangle_{(\sigma S_1)} |S_2 m_2\rangle. \quad (\text{A4})$$

Now, if the electron moves to site 2, we can couple $\boldsymbol{\sigma}$ and \mathbf{S}_2 to give $\bar{\mathbf{S}}_2$, which together with \mathbf{S}_1 yields another state of total spin S_T . However, as can be seen above, there are different ways to connect the three spins. One possible final state is

$$|\varphi_{\text{fi}}^A\rangle = |S_T m_T\rangle_{(S_1 (\sigma S_2))} \quad (\text{A5})$$

$$= \sum_{m_1 \bar{m}_2} \begin{pmatrix} S_1 & \bar{S}_2 & S_T \\ m_1 & \bar{m}_2 & m_T \end{pmatrix} |S_1 m_1\rangle |\bar{S}_2 \bar{m}_2\rangle_{(\sigma S_2)}. \quad (\text{A6})$$

The corresponding effective hopping matrix element $t_A^{(b)}$ is proportional to the overlap $\langle \varphi_{\text{fi}}^A | \varphi_{\text{in}} \rangle$, which can be evaluated by rewriting the multiple sum over the product of four Clebsch-Gordan coefficients with a 6- j symbol,

$$\langle \varphi_{\text{fi}}^A | \varphi_{\text{in}} \rangle = (-1)^{\bar{S}_1 + S_2 + S_T} \sqrt{(2\bar{S}_1 + 1)(2\bar{S}_2 + 1)} \times \begin{Bmatrix} \sigma & S_1 & \bar{S}_1 \\ S_T & S_2 & \bar{S}_2 \end{Bmatrix}. \quad (\text{A7})$$

We obtain the value of the matrix relevant for strong Hund’s coupling by setting $|\bar{S}_i| = S + 1/2 \equiv \bar{S}$ and $|S_i| = S = \bar{S} - 1/2$:

$$t_A^{(b)} = \frac{S_T + 1/2}{2\bar{S}} t, \quad (\text{A8})$$

i.e., there is no S_T -dependent phase factor. However, Müller-Hartmann and Dagotto derive an effective double-exchange Hamiltonian, where the hopping is expressed as the permutation of the spin \bar{S} “particle” at site 1 and the spin $(\bar{S} - 1/2)$ “hole” at site 2. Therefore the final state they have to consider for the matrix element is

$$|\varphi_{\text{fi}}^B\rangle = |S_T m_T\rangle_{((\sigma S_2) S_1)} \quad (\text{A9})$$

$$= \sum_{\bar{m}_2 m_1} \begin{pmatrix} \bar{S}_2 & S_1 & S_T \\ \bar{m}_2 & m_1 & m_T \end{pmatrix} |\bar{S}_2 \bar{m}_2\rangle_{(\sigma S_2)} |S_1 m_1\rangle. \quad (\text{A10})$$

Obviously the permutation of the indices in the Clebsch-Gordan coefficients yields a different phase factor in the overlap,

$$\langle \varphi_{\text{fi}}^B | \varphi_{\text{in}} \rangle = (-1)^{S_1 + S_2 + \bar{S}_1 + \bar{S}_2} \sqrt{(2\bar{S}_1 + 1)(2\bar{S}_2 + 1)} \times \begin{Bmatrix} \sigma & S_1 & \bar{S}_1 \\ S_T & S_2 & \bar{S}_2 \end{Bmatrix}, \quad (\text{A11})$$

and consequently the authors obtain a S_T -dependent phase factor for $t_B^{(b)}$,

$$t_B^{(b)} = (-1)^{2\bar{S}-S_T-1/2} \frac{S_T+1/2}{2\bar{S}} t. \quad (\text{A12})$$

To recover the effective Hamiltonian Eq. (6) of Ref. 8,

$$H^{\text{eff}} = -t P_{12} Q_{\bar{S}}(y), \quad (\text{A13})$$

we simply have to express S_T in $t_B^{(b)}$ with the help of $y = \mathbf{S}_1 \cdot \mathbf{S}_2 / [\bar{S}(\bar{S}-1/2)]$. In a direct way we can use interpolation polynomials in the form of Lagrange, i.e.,

$$Q_{\bar{S}}(y) = \sum_{S_T=1/2}^{2\bar{S}-1/2} t_B^{(b)} \prod_{\substack{j=1/2 \\ j \neq S_T}}^{2\bar{S}-1/2} \frac{y-y_j}{y_{S_T}-y_j} \quad (\text{A14})$$

$$y_j = \frac{j(j+1) - \bar{S}(\bar{S}+1) - (\bar{S}-1/2)(\bar{S}+1/2)}{2\bar{S}(\bar{S}-1/2)}, \quad (\text{A15})$$

or the recursive formula given in Eq. (7) of Ref. 8. Of course, taking the matrix element $t_A^{(b)}$ for the construction of $Q_{\bar{S}}(y)$ is wrong, as this does not account for the phase factor due to the permutation P_{12} . Concerning the limit of classical spins, we can see no connection between the above S_T -dependent phase factor and the Berry phase.

The peculiarities concerning the phase factor indicate that the above derivation is not suitable for the generalization to a lattice. Moreover, the spin- \bar{S} ‘‘particles’’ still obey fermion commutation relations, which cannot be expressed by conventional permutation or spin operators. That means, the expression given in Eq. (6) of Ref. 8,

$$H^{\text{eff}} = -t \sum_{\langle ij \rangle} P_{ij} Q_{\bar{S}}(y), \quad (\text{A16})$$

is *not* the correct quantum DE Hamiltonian on a lattice.

APPENDIX B: CLASSICAL LIMIT OF THE DE MODEL

The limit $S \rightarrow \infty$ of $H_{\text{el}}^{\text{DE}}$, Eq. (15), is easily derived by taking its expectation value with spin coherent states,¹⁰

$$|\Omega(S, \theta, \phi)\rangle = \frac{(ua^\dagger + vb^\dagger)^{2S}}{\sqrt{(2S)!}} |0\rangle, \quad (\text{B1})$$

where $u = \cos(\theta/2)e^{i\phi/2}$ and $v = \sin(\theta/2)e^{-i\phi/2}$. Using the properties of coherent states,

$$a|\Omega(S, \theta, \phi)\rangle = \sqrt{2Su}|\Omega(S-\frac{1}{2}, \theta, \phi)\rangle, \quad (\text{B2})$$

$$b|\Omega(S, \theta, \phi)\rangle = \sqrt{2Sv}|\Omega(S-\frac{1}{2}, \theta, \phi)\rangle, \quad (\text{B3})$$

for a given spin configuration $\{\theta_k, \phi_k\}$ and two electronic states $|\psi_1\rangle$ and $|\psi_2\rangle$,

$$|\psi_j\rangle = \prod_k |n_{j,k}\rangle \left| \Omega \left(S + \frac{n_{j,k}}{2}, \theta_k, \phi_k \right) \right\rangle \quad (\text{B4})$$

[where $|n_{j,k}\rangle = (c_k^\dagger)^{n_{j,k}}|0\rangle$ with numbers $n_{j,k} \in \{0,1\}$], we find the average

$$\langle \psi_1 | H_{\text{el}}^{\text{DE}} | \psi_2 \rangle = \prod_k \langle n_{1,k} | \left(- \sum_{\langle ij \rangle} [t_{ij} c_i^\dagger c_j + \text{H.c.}] \right) \prod_k |n_{2,k}\rangle \quad (\text{B5})$$

with the matrix element

$$t_{ij} = t \left[\cos\left(\frac{\theta_i}{2}\right) \cos\left(\frac{\theta_j}{2}\right) e^{-i(\phi_i - \phi_j)/2} + \sin\left(\frac{\theta_i}{2}\right) \sin\left(\frac{\theta_j}{2}\right) e^{i(\phi_i - \phi_j)/2} \right]. \quad (\text{B6})$$

Hence, the classical Hamiltonian should read

$$H_{\text{class}}^{\text{DE}} = - \sum_{\langle ij \rangle} [t_{ij} c_i^\dagger c_j + \text{H.c.}], \quad (\text{B7})$$

which is equivalent to the results obtained in Refs. 11 and 8. In our case, however, the classical limit followed from the quantum Hamiltonian, Eq. (15), in an obvious and more straightforward way. Note also, that averaging the operators $(R_i^+)^{\dagger}$ and R_i^+ over coherent states yields the unitary transformation onto rotated electrons $d_j^{(\dagger)}$ (compare Refs. 11 and 8),

$$d_j = \cos\left(\frac{\theta_j}{2}\right) e^{-i\phi_j/2} \tilde{c}_{j\uparrow} + \sin\left(\frac{\theta_j}{2}\right) e^{i\phi_j/2} \tilde{c}_{j\downarrow}, \quad (\text{B8})$$

i.e., naturally Eq. (10) has the same classical limit.

To check whether the description of double-exchange in terms of classical spins is appropriate, we compared the (canonical) density of states (DOS) for a fixed number of carriers on a small cluster, which interact with quantum [Eq. (15)] or classical [Eq. (B7)] spins, respectively. Using Chebyshev expansion and maximum entropy methods,¹² for

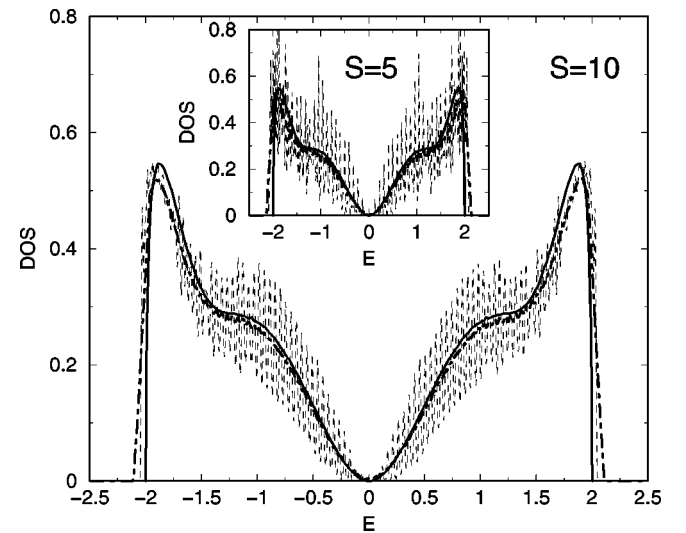


FIG. 1. Density of nonzero eigenvalues (dashed line) and running average (bold dot-dashed line), calculated for two electrons on four sites with $S=5$ (inset) and $S=10$, compared to the classical result $S \rightarrow \infty$ (bold solid line).

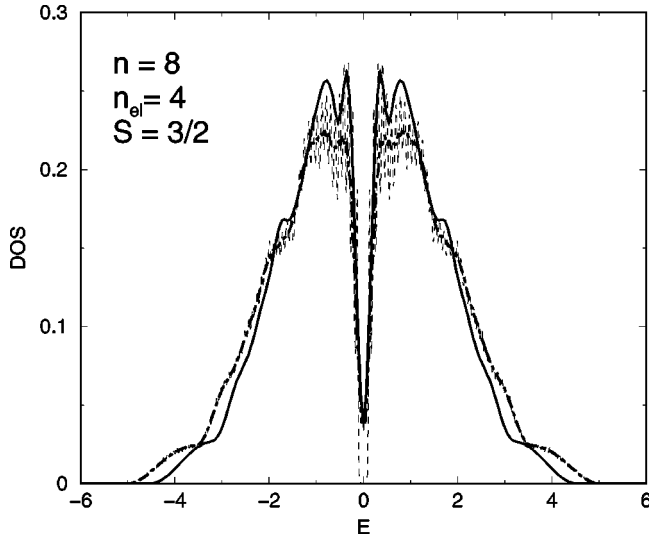


FIG. 2. Density of nonzero eigenvalues for four electrons on eight sites with $S=3/2$; same notations as in Fig. 1.

the quantum case the spectra can be obtained numerically for rather large S . The classical DOS is found by averaging the eigenvalues of $H_{\text{class}}^{\text{DE}}$, Eq. (B7), over a large number of spin configurations.

In Fig. 1 we consider two electrons on a ring of four sites. Comparing the running average (bold dot-dashed line) over the discrete spectrum (thin dashed line) and the classical limit (bold solid line), we find good convergence already for a moderate spin length $S=10$. But even for the case $S=3/2$, which is realized in the manganites, the classical description appears to be acceptable. If we consider four electrons on a ring of eight sites, the spectrum is much more dense, allowing similarities to be recognized easily, see Fig. 2. Of course, from the density of states we can learn nothing about correlations or other more involved features. Note that in both figures we subtracted the peak at $E=0$ consuming a large fraction of spectral weight.

Another interesting check concerns the effective hopping matrix element $\tilde{t}^{(b)}$, Eq. (26). With classical spins and the

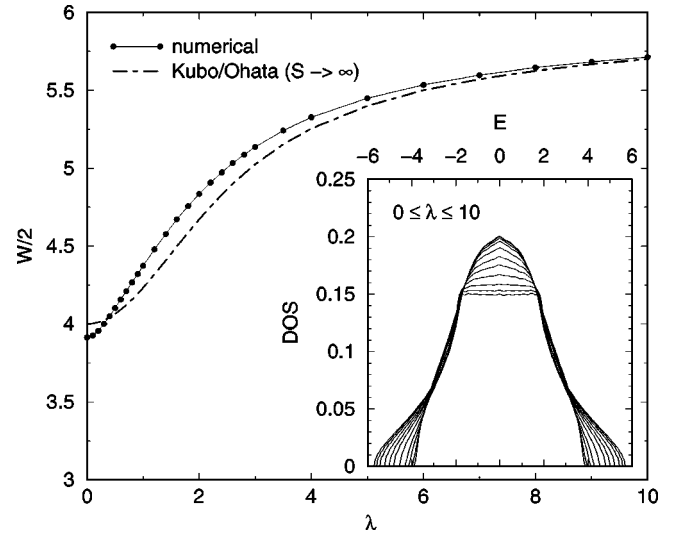


FIG. 3. Bandwidth of tight-binding electrons on a simple cubic lattice of size 64^3 , interacting with thermalized classical spins in a homogeneous field λ , compared to the limit $S \rightarrow \infty$ of the Kubo-Ohata formula. Inset: Narrowing of the corresponding density of states with decreasing λ .

Chebyshev expansion methods mentioned above, one can calculate the grand-canonical DOS of the tight-binding model, Eq. (B7), for rather large clusters (here 64^3 sites on a simple cubic lattice) without much effort. By considering a thermalized ensemble of classical spins in a homogeneous field we compare the resulting bandwidth with the limit $S \rightarrow \infty$ of $\tilde{t}^{(b)}$, where the limit of $\gamma_{\bar{S}}[\bar{S}\lambda]$ is obtained omitting \bar{S} in the argument and setting $\bar{S} \rightarrow \infty$ in the index,

$$\gamma_{\bar{S} \rightarrow \infty}[\lambda] = \frac{1}{2} \left(1 + \coth(\lambda) \left[\coth(\lambda) - \frac{1}{\lambda} \right] \right). \quad (\text{B9})$$

The agreement is rather satisfactory, as can be seen in Fig. 3. Naturally, the precise shape of the DOS (see inset) will not be reproduced by the effective electronic model; it remains simple cubic tight-binding for all fields and temperatures.

¹C. Zener, Phys. Rev. **82**, 403 (1951).

²S. Jin, T.H. Tiefel, M. McCormack, R.A. Fastnach, R. Ramesh, and L.H. Chen, Science **264**, 413 (1994).

³P.W. Anderson and H. Hasegawa, Phys. Rev. **100**, 675 (1955).

⁴A. Weiße, J. Loos, and H. Fehske, cond-mat/0101235 (unpublished).

⁵S.K. Sarker, J. Phys.: Condens. Matter **8**, L515 (1996); D.P. Arovas and F. Guinea, Phys. Rev. B **58**, 9150 (1998); S. Ishizaka and S. Ishihara, *ibid.* **59**, 8375 (1999); R. Kilian, Ph.D. thesis, Technische Universität Dresden, 1999; K. Penc and R. Lacaze, Europhys. Lett. **48**, 561 (1999); R.Y. Gu, Z.D. Wang, S.-Q. Shen, and D.Y. Xing, Phys. Rev. B **61**, 1211 (2000).

⁶A.E. Bocquet, T. Mizokawa, T. Saitoh, H. Namatame, and A. Fujimori, Phys. Rev. B **46**, 3771 (1992); J. Zaanen and G.A.

Sawatzky, J. Solid State Chem. **88**, 8 (1990); T. Mizokawa and A. Fujimori, Phys. Rev. B **51**, 12 880 (1995).

⁷K. Kubo and N. Ohata, J. Phys. Soc. Jpn. **33**, 21 (1972).

⁸E. Müller-Hartmann and E. Dagotto, Phys. Rev. B **54**, R6819 (1996).

⁹A. Lindner, *Drehimpulse in der Quantenmechanik* (Teubner, Stuttgart, 1984).

¹⁰A. Auerbach, *Interacting Electrons and Quantum Magnetism* (Springer-Verlag, New York, 1994).

¹¹E.M. Kogan and M.I. Auslender, Phys. Status Solidi B **147**, 613 (1988).

¹²R.N. Silver, H. Röder, A.F. Voter, and J.D. Kress, J. Comput. Phys. **124**, 115 (1996); R.N. Silver and H. Röder, Phys. Rev. E **56**, 4822 (1997); B. Bäuml, G. Wellein, and H. Fehske, Phys. Rev. B **58**, 3663 (1998).