Enhanced ferromagnetism from electron-electron interactions in double-exchange-type models

Nuri A. Yazdani and Malcolm P. Kennett

Department of Physics, Simon Fraser University, 8888 University Drive, Burnaby, British Columbia, V5A 1S6 Canada
(Received 27 January 2011; published 6 April 2011)

The magnetic properties of a variety of materials with promise for technological applications have been described by models in which fermions are coupled to local moment spins. Monte Carlo studies of such models usually ignore electron-electron interactions, even though the energy scale corresponding to these interactions may be comparable to or larger than other relevant energy scales. In this work we add on-site interactions between fermions to the double-exchange model which we study with a Monte Carlo scheme in which temporal fluctuations of local moment spins are fully accounted for and electron-electron interactions are treated at a mean-field level. We show that when the number of fermions is considerably less than the number of local moments even moderate interactions can lead to significant enhancement of ferromagnetism and the Curie temperature.

I. INTRODUCTION

The magnetic properties of a variety of materials of fundamental interest and technological promise, such as colossal magnetoresistance (CMR) manganites,[1] rare-earth hexaborides,[2] and diluted, magnetic semiconductors (DMS),[3] have been described using double-exchange- (DE) type models in which fermions are coupled to local moment spins. In the limit that the number of fermions is considerably less than the number of local moments, such models generally display ferromagnetism.[4]

In manganites it is well established that electron-electron interactions are at least as large as the Hund coupling and may be the largest energy scale in the problem.[1,5–7] The importance of interactions has also been stressed for DMS[8–10] and hexaborides.[11] The combination of disorder and electron-electron interactions may also play an important role in nanoscale electronic phase separation,[12] which has been argued to be important for CMR in manganites.[13] It is hence important to develop accurate techniques that can account for the effects of electron-electron interactions in DE-type models and to study their effects on magnetic properties of these models.

Here we introduce a method to study the effects of electron-electron interactions in DE-type models when the energy scale for interactions is in the experimentally relevant regime of no more than a few times the Hund coupling. We use this method to show that even moderate interactions can enhance ferromagnetism and the Curie temperature T_c significantly when the number of fermions is considerably less than the number of local moments.

We combine a Hartree-Fock treatment of electron-electron interactions with a Monte Carlo scheme for fermions coupled to classical local moment spins. Through the use of exact diagonalizations on small systems we have determined regions of parameter space where this technique should be most accurate. We calculate the magnetization as a function of temperature for a variety of interaction strengths and determine the effects of interactions on the Curie temperature T_c. Electron-electron interactions can lead to ferromagnetism in the absence of Hund coupling,[14] so the enhancement in magnetization that we find can be understood as this tendency reinforcing the ferromagnetism that arises from the Hund coupling.

II. MODEL AND MONTE CARLO SCHEME

The general Hamiltonian we consider is of the form

$$\mathcal{H} = \mathcal{H}_{\mathrm{DEM}} + \mathcal{H}_{\mathrm{int}}, \qquad (1)$$

with

$$\mathcal{H}_{\mathrm{DEM}} = -\sum_{ij}[t_{ij}c_{i\sigma}^{\dagger}c_{j\sigma} + \mathrm{H.c.}] + \sum_{ij} J_{ij}\mathbf{S}_i \cdot \mathbf{s}_i,$$

$$\mathcal{H}_{\mathrm{int}} = U \sum_i n_{i\uparrow}n_{i\downarrow},$$

where i and j are site indices, t_{ij} is the hopping integral, J_{ij} is the Hund coupling, U is the on-site Hubbard repulsion, $c_{i\sigma}^{\dagger}$ creates a fermion with spin σ on site i, $\mathbf{S}_i$ is a local moment spin on site i, and $\mathbf{s}_j = \frac{1}{2} \sum_{\alpha,\beta}(c_{j\alpha}^{\dagger}\boldsymbol{\sigma}_{\alpha\beta}c_{j\beta})$ is the fermion spin on site j. For simplicity, we assume the hopping is only to nearest-neighbor sites on a cubic lattice with amplitude $t_{ij} = t$ and that the Hund coupling is purely local: $J_{ij} = J\delta_{ij}$. We assume that there is a local moment on every site in the lattice and that the fermions have spin-$\frac{1}{2}$ (these assumptions can be easily relaxed).

There has been some work to study the effects of finite U in models of the form Eq. (1) using exact diagonalizations,[15] DMRG,[16] and mean-field approximations.[9] However, these techniques are not appropriate for studying finite temperature magnetic properties in dimensions higher than one taking into account the temporal fluctuations of spins. In order to determine the finite temperature magnetic properties of the Hamiltonian Eq. (1), it is necessary to perform Monte Carlo simulations. If there are N local moment spins with spin S, then the size of the Hilbert space scales as $(2S + 1)^N$, hence it is usual to approximate the local moment spin as classical, which is often reasonable, given that in many systems of interest S is larger than $\frac{1}{2}$. Previous such Monte Carlo simulations[8,17–20] have restricted their attention to models of the form Eq. (1) with $U = 0$ with the exception of Ref. 8. In Ref. 8 interactions were included for some parameter values using a zero-temperature variational procedure, due to convergence issues with Hartree-Fock, and did not appear to have a strong influence on magnetic properties. The model in Ref. 8 also included disorder and spin-orbit coupling, and these

terms may have influenced the convergence of Hartree-Fock calculations.

We write the classical local moment spins in the form $\mathbf{S}_i = (S_i^z, \phi_i)$, and for a specific arrangement of local moment spins, one can diagonalize Eq. (1) with $U = 0$:

$$\mathcal{H}(\{\mathbf{S}_i\}) = \sum_m E_m(\{\mathbf{S}_i\}) a_m^\dagger a_m, \quad (2)$$

where $\{E_m(\{\mathbf{S}_i\})\}$ are the eigenvalues for a given local moment spin configuration. $a_m^\dagger$ and a_m are the creation and annihilation operators for the mth eigenstate of H:

$$a_m^\dagger = \sum_{i\sigma} \psi_{im\sigma} c_{i\sigma}^\dagger, \quad a_m = \sum_{i\sigma} \psi_{im\sigma}^* c_{i\sigma}.$$

This allows one to write the fermion free energy as

$$\mathcal{F}(\{\mathbf{S}_i\}) = -\frac{1}{\beta} \sum_{m=1}^{2N} \ln\{1 + e^{-\beta[E_m(\{\mathbf{S}_i\}) - \mu]}\}, \quad (3)$$

where $\beta = 1/k_B T$ is the inverse temperature and hence the classical partition function for the N classical spins takes the form

$$Z = \left[\prod_{i=1}^N \int_{-1}^1 dS_i^z \int_0^{2\pi} d\phi_i \right] e^{-\beta \mathcal{F}(\{\mathbf{S}_i\})}. \quad (4)$$

Casting the partition function in this form makes it clear that we may use the Metropolis algorithm to determine whether to flip a spin, with the change in the fermion free energy determining whether a spin flip is accepted or rejected in the classical spin Monte Carlo simulation.

Introducing a finite U greatly increases the size of the fermion Hilbert space: For N local moments and n fermions there are $(2N)!/(2N - n)!n!$ states, as compared to $2N$ noninteracting states. This renders even moderate values of N out of reach computationally. In order to explore larger values of N the interaction term must be treated in an approximate fashion, hence our use of Hartree-Fock as the simplest self-consistent approach. The Hartree-Fock approximation reduces the size of the fermion Hilbert space to $2N$ and allows for the use of the Monte Carlo scheme outlined above, with Hartree-Fock energies replacing the $E_m(\{\mathbf{S}_i\})$.

We decompose H_{int} in the Hamiltonian Eq. (1) as

$$U \sum_i n_{i\uparrow} n_{i\downarrow} \simeq U \sum_i [\langle n_{i\uparrow}\rangle c_{i\downarrow}^\dagger c_{i\downarrow} + \langle n_{i\downarrow}\rangle c_{i\uparrow}^\dagger c_{i\uparrow}$$
$$- \langle c_{i\uparrow}^\dagger c_{i\downarrow}\rangle c_{i\downarrow}^\dagger c_{i\uparrow} - \langle c_{i\downarrow}^\dagger c_{i\uparrow}\rangle c_{i\uparrow}^\dagger c_{i\downarrow}$$
$$- \langle n_{i\uparrow}\rangle\langle n_{i\downarrow}\rangle + \langle c_{i\uparrow}^\dagger c_{i\downarrow}\rangle\langle c_{i\downarrow}^\dagger c_{i\uparrow}\rangle]. \quad (5)$$

Approximating H_{int} with Eq. (5) we can write

$$H \simeq H_{\text{HF}} - U \sum_i [\langle n_{i\uparrow}\rangle\langle n_{i\downarrow}\rangle - \langle c_{i\uparrow}^\dagger c_{i\downarrow}\rangle\langle c_{i\downarrow}^\dagger c_{i\uparrow}\rangle],$$

and the single-particle states satisfy

$$H_{\text{HF}}|\phi_m\rangle = \epsilon_m|\phi_m\rangle,$$

with single-particle energies

$$\tilde{\epsilon}_m = \epsilon_m - \frac{U}{2} \sum_i [\langle n_{i\uparrow}\rangle \psi_{im\uparrow}^* \psi_{im\uparrow} + \langle n_{i\downarrow}\rangle \psi_{im\downarrow}^* \psi_{im\downarrow}$$
$$- \langle c_{i\uparrow}^\dagger c_{i\downarrow}\rangle \psi_{im\downarrow}^* \psi_{im\uparrow} - \langle c_{i\downarrow}^\dagger c_{i\uparrow}\rangle \psi_{im\uparrow}^* \psi_{im\downarrow}], \quad (6)$$

which we calculate by iterating to self-consistency and use to determine the approximate fermion free energy for each local moment spin configuration $\{\mathbf{S}_i\}$.

III. NUMERICAL RESULTS

A. Exact diagonalization

The Hartree-Fock approximation is uncontrolled, hence in order to ascertain the regions of parameter space in which the hybrid Hartree-Fock Monte Carlo scheme should be most accurate, we performed exact diagonalization calculations on eight site $2 \times 2 \times 2$ systems with two, three, and four electrons and compared the energies of the exact ground state, first excited state, and second excited state averaged over 25 different local moment configurations. In Fig. 1 we show the relative error $\Delta_E = |E_n - E_n^{\text{HF}}|/|E_n|$ as a function of J/t and U/t, where E_n is the exact energy and E_n^{HF} is the Hartree-Fock approximation to the energy.

Provided U is not too large in comparison to J, the Hartree-Fock approximation gives a good account of the low-lying energy levels, and we found that for $U \lesssim 3J$ the relative error was less than 5% with the error decreasing with decreased U/J, hence we restricted our Monte Carlo simulations to this region. We additionally checked the relative error in the densities between the exact diagonalization results and the Hartree-Fock approximation and found the best agreement in the same region of parameter space as for the energy.
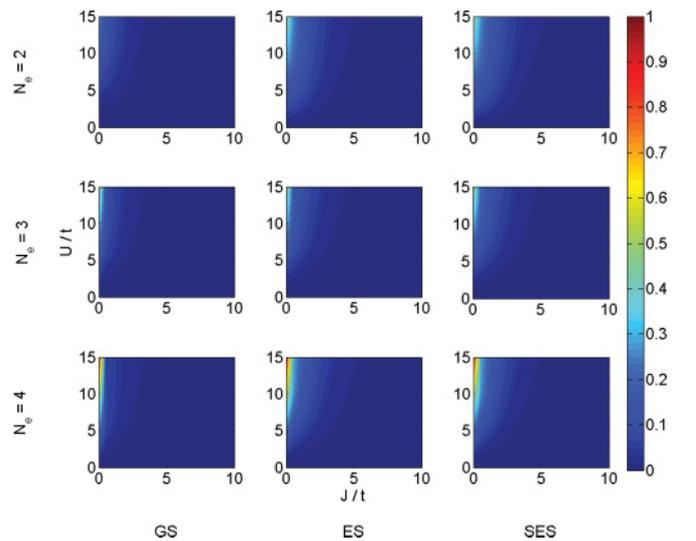

FIG. 1. (Color) Relative error between the exact and the Hartree-Fock evaluation of the ground state, first excited state, and second excited state energies for an eight-site system, with two, three, and four electrons, averaged over 25 local moment configurations, as a function of U/t and J/t.
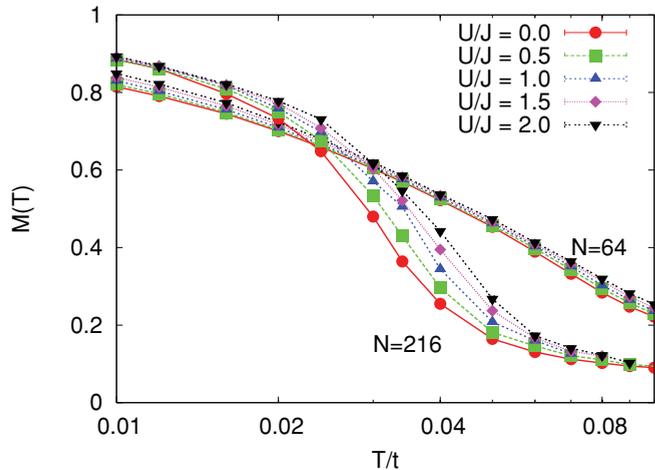
FIG. 2. (Color online) Local moment magnetization for $J/t = 5$ for $N = 64$ and 216 local moments for $U/J = 0.0$, 0.5, 1.0, 1.5, and 2.0.

B. Monte Carlo simulations

We performed Monte Carlo simulations for cubic systems with $N = 4^3 = 64$ and $N = 6^3 = 216$ local moments, with $n = 8$ and $n = 27$ fermions respectively corresponding to $n/N = 1/8$ in both cases. These system sizes are competitive with recent simulations in noninteracting systems.[20] We used a similar equilibration procedure to Ref. 19, by bringing two replicas of the system into equilibrium, evolving under Metropolis dynamics, with an additional self-consistent Hartree-Fock loop as discussed above. We used the z test[21] to determine when there was 95% confidence that the two replicas were in equilibrium and then collected data for a further 10 000 Monte Carlo sweeps.

We calculated the magnetization of the local moments

$$M(T) = \left\langle \frac{1}{NS}\sqrt{\left|\sum_i \mathbf{S}_i\right|^2}\right\rangle,$$

as a function of temperature, where the angle brackets indicate a thermodynamic average. In Fig. 2 we show the local moment magnetization when $J/t = 5$ calculated both for $N = 64$ and $N = 216$ local moments. There is some enhancement of the magnetization with increasing U/J in the $N = 64$ samples. However, the enhancement is much clearer in the $N = 216$ data, where the finite size effects that enhance the magnetization at higher temperatures in the $N = 64$ data are much smaller, and the magnetization curve is more reflective of the thermodynamic limit. Calculations of the fermion magnetization yield similar enhancement with increasing interaction strength.[22]

Due to the evident finite size effects in the magnetization, we use the Binder cumulant[23]

$$g(N,T) = \frac{1}{2}\left[5 - 3\left(\frac{\langle M^4\rangle}{\langle M^2\rangle^2}\right)\right],$$

to determine T_c as a function of interaction strength, as $g(N,T)$ should be independent of system size at T_c. Our results for $J/t = 5$, determined using the crossing point of Binder cumulant curves for $N = 64$ and $N = 216$ are displayed in
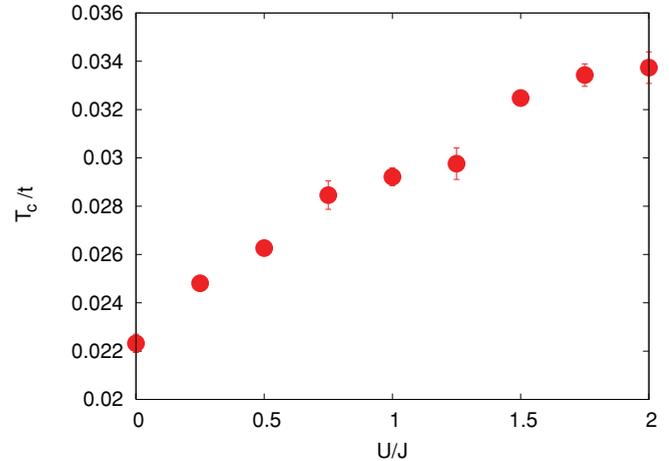

FIG. 3. (Color online) Curie temperature as a function of U/J for $J/t = 5$.

Fig. 3. There is a monotonic increase in T_c with increasing U/J, with an almost 50% increase in T_c between $U/J = 0$ and $U/J = 2$.

IV. CONCLUSIONS

By performing Monte Carlo simulations of an interacting DE model, we have demonstrated that electron-electron interactions, which are known or expected to be important in a variety of materials whose magnetic properties are described by DE-type models, can lead to quantitatively important increases of the Curie temperature. The Hubbard model can display ferromagnetism,[14] hence it is natural that in the presence of Hund coupling, which independently promotes ferromagnetism, the two terms in the Hamiltonian can reinforce each other to produce the the approximately linear in U enhancement observed here. As a mean-field approximation, Hartree-Fock will tend to overestimate the tendency to ordering as U/J is increased, but the effects observed here should be robust to fluctuations. This is because the interactions enhance an existing tendency to ferromagnetism rather than imposing order on an otherwise disordered system. Further, our use of Hartree-Fock is confined to values of U/J where exact diagonalizations suggest that it will be most accurate.

The scheme we have introduced, and our demonstration that it should be accurate for the experimentally relevant interaction range $U \lesssim 3J$, should be a further step toward the quantitative description of the magnetic properties of important DE-type materials. It should also be possible to extend the scheme introduced here to treat both disorder (which has been included in previous noninteracting simulations[8,18–20]) and long-range Coulomb interactions, which have been argued as being relevant to nanoscale phase separation and CMR.[12]

[1] E. Dagotto, T. Hotta, and A. Moreo, Phys. Rep. **344**, 1 (2001).

[2] V. M. Pereira, J. M. B. Lopesdos Santos, E. V. Castro, and A. H. Castro-Neto, Phys. Rev. Lett. **93**, 147202 (2004).

[3] T. Dietl *et al.*, Science **287**, 1019 (2000).

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

[5] T. V. Ramakrishnan, J. Phys. Cond. Mat. **19**, 125211 (2007); K. H. Ahn and A. J. Millis, J. Appl. Phys. **87**, 5013 (2000).

[6] S. Satpathy, Z. S. Popovic, and F. R. Vukajlovic, Phys. Rev. Lett. **76**, 960 (1996); J.-H. Park, C. T. Chen, S.-W. Cheong, W. Bao, G. Megis, V. Chakraian, and Y. U. Idzerda, *ibid.* **76**, 4215 (1996).

[7] D. I. Golosov, Phys. Rev. B **71**, 014428 (2005); Phys. Rev. Lett. **104**, 207207 (2010).

[8] G. A. Fiete, G. Zarand, K. Damle, and C. P. Moca, Phys. Rev. B **72**, 045212 (2005).

[9] M. Berciu and R. N. Bhatt, Phys. Rev. B **69**, 045202 (2004).

[10] T. Jungwirth, W. A. Atkinson, B. H. Lee, and A. H. MacDonald, Phys. Rev. B **59**, 9818 (1999); S.-R. Eric Yang and A. H. MacDonald, *ibid.* **67**, 155202 (2003); J. Ohe *et al.*, J. Phys. Soc. Jpn. **78**, 083703 (2009).

[11] V. M. Pereira, J. M. B. Lopes dos Santos, and A. H. Castro Neto, e-print arXiv:0804.3094v1.

[12] V. B. Shenoy, T. Gupta, H. R. Krishnamurthy, and T. V. Ramakrishnan, Phys. Rev. B **80**, 125121 (2009).

[13] J. Burgy, M. Mayr, V. Martin-Mayor, A. Moreo, and E. Dagotto, Phys. Rev. Lett. **87**, 277202 (2001).

[14] J. Hubbard, Proc. Roy. Soc. Lond. A **276**, 238 (1963).

[15] S. Mishra, R. Pandit, and S. Satpathy, J. Phys. Cond. Mat. **11**, 8561 (1999).

[16] A. L. Malvezzi, S. Yunoki, and E. Dagotto, Phys. Rev. B **59**, 7033 (1999).

[17] S. Yunoki, J. Hu, A. L. Malvezzi, A. Moreo, N. Furukawa, and E. Dagotto, Phys. Rev. Lett. **80**, 845 (1998); M. J. Calderon and L. Brey, Phys. Rev. B **58**, 3286 (1998); Y. Motome and N. Furukawa, J. Phys. Soc. Jpn. **68**, 3853 (1999); A. Moreo, S. Yunoki, and E. Dagotto, Science **283**, 2034 (1999); J. L. Alonso *et al.*, Nucl. Phys. B **596**, 587 (2000).

[18] J. Schliemann, J. König, and A. H. MacDonald, Phys. Rev. B **64**, 165201 (2001); R. N. Bhatt *et al.*, J. Supercond. **15**, 71 (2002); Comp. Phys. Commun. **147**, 684 (2002); M. Mayr, G. Alvarez, and E. Dagotto, Phys. Rev. B **65**, 241202(R) (2002); M. J. Calderón, G. Gómez-Santos, and L. Brey, *ibid.* **66**, 075218 (2002); M. P. Kennett *et al.*, J. Magn. Magn. Mat. **272-276**, 1993 (2004); G. A. Fiete, G. Zarand, B. Janko, P. Redlinski, and C. P. Moca, Phys. Rev. B **71**, 115202 (2005); Y. Yildirim, G. Alvarez, A. Moreo, and E. Dagotto, Phys. Rev. Lett. **99**, 057207 (2007).

[19] M. P. Kennett, M. Berciu, and R. N. Bhatt, Phys. Rev. B **66**, 045207 (2002).

[20] G. Alvarez and E. Dagotto, Phys. Rev. B **68**, 045202 (2003).

[21] R. C. Sprinthall, *Basic Statistical Analysis*, 7th ed. (Pearson, New York, 2003).

[22] N. A. Yazdani and M. P. Kennett (in preparation).

[23] K. Binder and A. P. Young, Rev. Mod. Phys. **58**, 801 (1986).