Metallic ferromagnetism in a single-band model: Effect of band filling and Coulomb interactions

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A single-band tight-binding model with on-site repulsion and nearest-neighbor exchange interaction has been proposed as a simple model to describe metallic ferromagnetism. Here we extend previously obtained exact-diagonalization studies for a one-dimensional 1/2-filled band system to other band fillings, and consider the effect of including various other Coulomb matrix elements in the Hamiltonian that are expected to be of appreciable magnitude in real materials. Results of exact diagonalization and mean-field theory for the one-dimensional case are compared. As the band filling decreases from 1/2, the tendency to ferromagnetism is found to decrease in exact diagonalization, while mean-field theory predicts the opposite behavior. A nearest-neighbor Coulomb repulsion term is found to suppress the tendency to ferromagnetism; however, the effect becomes small for large on-site repulsion. A pair hopping interaction enhances the tendency to ferromagnetism. A nearest-neighbor hybrid Coulomb matrix element breaks electron-hole symmetry and causes metallic ferromagnetism to occur preferentially for more than half-filled rather than less-than-half-filled bands in this model. Mean-field theory is found to yield qualitatively incorrect results for the effect of these interactions on the tendency to ferromagnetism. The implications of these results for the understanding of ferromagnetism in real materials is discussed. [S0163-1829(96)03733-2]

I. INTRODUCTION

The Hubbard model was originally proposed as a simple model to describe the physics of metallic ferromagnetism.[1] It gives rise to ferromagnetism for large values of the on-site Coulomb repulsion U within mean-field theory[2] (where it is equivalent to the Stoner model,[3]) as well as within other approximations.[1,4] However, subsequent work has shown that an on-site Coulomb repulsion U by itself will not give rise to metallic ferromagnetism[5,6] except in special situations, such as a single hole in a half-filled band[7] or special lattice geometries.[8,9] That is, it appears that in general electrons of antiparallel spin can more easily avoid paying the price of on-site Coulomb repulsion by developing spatial correlations rather than by spin polarizing, contrary to the predictions of mean-field theory. This then leads to the question of what is the simplest model beyond the Hubbard model that contains the essential physics of metallic ferromagnetism. Two natural ways to go beyond the original Hubbard model are to include band degeneracy, which allows for intra-atomic exchange, or to include other Coulomb interaction matrix elements within the single-band model. We explore the latter one in this work.

The possible importance of ''off-diagonal'' Coulomb matrix elements in tight-binding models was pointed out by Kivelson *et al.*,[10] in the context of attempting to understand the effect of Coulomb interactions on the Peierls instability in one-dimensional (1D) metals. One of these off-diagonal matrix elements is the Coulomb exchange integral J. Even though Heisenberg had considered the effect of the nearest-neighbor exchange integral J on ferromagnetism back in 1928,[11] the role of this parameter in *metallic* ferromagnetism remained unexplored until recently. This may be due to the fact that J represents the quantum-mechanical exchange energy for *localized* rather than for itinerant electrons (which was the physical picture behind Heisenberg's work), and the fact that there are other Coulomb interaction terms that are much larger in magnitude than J that play a dynamic role in itinerant electron systems, such as the on-site repulsion U and the nearest-neighbor repulsion V. As a consequence, the effect of J on ferromagnetism had only been considered for insulators in the past.

Recently we began a study of the problem of metallic ferromagnetism under the assumption that the nearest-neighbor exchange interaction J, even if small in magnitude, may play a central role in it.[12–17] This was based on the observation that J lowers the direct repulsion energy of an itinerant electron in a bonding and another in an antibonding state, a situation that arises when spin polarization occurs, particularly for a half-filled band. Since this could not occur for free electrons in a continuum, it was argued that it is the combination of the ionic lattice potential and the electronic Coulomb repulsion that is responsible for metallic ferromagnetism. In fact, the possibility that ferromagnetism could be helped by lowering of the direct Coulomb energy on repopulation of the k states, and that this effect may be appreciable for very compact Wannier functions, had been suggested in the past by Herring[18] and by Wohlfarth.[19]

A mean-field solution of a Hamiltonian with interactions U and J revealed several interesting features of the model. It was found that for large U ferromagnetism would occur for rather small values of J. However, the qualitative features of the theory were dictated by J rather than by U; in other words, the model with $J=0$ (Stoner model) is qualitatively different to the one with any $J\neq 0$. The mean-field treatment also showed that a driving force for metallic ferromagnetism is the energy increase due to band narrowing (effective mass enhancement) that occurs as the temperature is lowered in the presence of the interaction J, which is suppressed when spin polarization develops. Within this picture then it is not the competition between kinetic and potential energy that determines whether the nonmagnetic or the ferromagnetic

state is favored, as commonly understood, but instead ferromagnetism itself should be understood as ''kinetic energy driven.'' Various features of this model found within mean-field theory that are in agreement with experimental observations are discussed in Refs. 12–14.

The most favorable situation for ferromagnetism in the model with interactions U and J was found to be the half-filled band case, as expected from qualitative arguments. In fact, Campbell *et al.*[20] had found earlier that in a one-dimensional half-filled band ferromagnetism would occur in the presence of a large nearest-neighbor exchange (termed a ''bond-charge repulsion'' in that work). We explored that case in more detail by exact diagonalization of a one-dimensional model,[15] and studied the stability of the half-filled fully polarized ferromagnetic state in d dimensions by exact solution of the problem of one overturned spin.[15,16] These exact results were found to be in qualitative and semiquantitative agreement with the results of mean-field theory. In particular, they showed that for small U the system with one overturned spin is metallic, corresponding to unbound down-spin electron and up-spin hole, and that in this regime the magnetization decreases continuously from the fully polarized state; in contrast, for large U, the down-spin electron and up-spin hole are bound, the system is insulating, and the magnetization jumps discontinuously from the fully polarized state. van Dongen and Janis[21] have suggested that this transition from metallic to insulating state in the almost fully polarized subspace is a precursor of the Mott-Hubbard transition in the unpolarized state.

Recently, Strack and Vollhardt[22] have derived exact criteria for the existence of ferromagnetism in generalized tight-binding models at half-filling and also concluded that the nearest-neighbor exchange interaction J plays a fundamental role. Furthermore, they extended their study to the case of one hole in a half-filled band,[23] previously studied by Nagaoka[7] for the Hubbard model and found that also in that case the nearest-neighbor exchange is important for stabilizing ferromagnetism at finite U. Campbell and co-workers[24] studied a one-dimensional half-filled generalized tight-binding model by a variety of techniques. Even though the focus of that work was on the tendency to lattice dimerization, they also established that in a wide region of parameter space ferromagnetism would occur in the presence of the interaction J.

In this paper we explore the effects of different band fillings and of other interaction parameters by exact diagonalization of a one-dimensional model, by mean-field theory, and by exact solution of the Schrödinger equation for simple limiting cases. Unlike our previous findings, it is found that the mean-field theory predictions are in qualitative *disagreement* with exact results when we consider band fillings other than 1/2 and the effect of other interactions. Generally, it is found that mean-field theory severely overestimates the tendency to ferromagnetism. In particular, for band fillings other than 1/2 we find that a finite value of J is needed for ferromagnetism even in the limit $U\rightarrow\infty$. Nevertheless, the results found here support the conjecture[12] that the nearest-neighbor exchange interaction J plays a fundamental role in itinerant ferromagnetism.

Because metallic ferromagnetism has been found in nature so far only in systems with degenerate electron bands (d or f), it is commonly believed that band degeneracy, which in particular allows for the existence of intra-atomic exchange, is essential for its existence. However, in a material such as Ni where there is less than one hole per d-shell atom, it is not likely that either band degeneracy or intra-atomic exchange play any role. For that case, a model such as the one discussed here would appear to be qualitatively appropiate. For other materials where intra-atomic exchange should play a role (such as Fe), it is possible that such role is quantitatively important but does not alter the physics of the single-band model considered here in an essential way.

This paper is organized as follows: In Sec. II we define the model and discuss various analytic results. Section III gives numerical results for the one-dimensional model, and we conclude in Sec. IV with a discussion.

II. MODEL AND SOME ANALYTIC RESULTS

A single-band tight-binding Hamiltonian with all Coulomb matrix elements included is given by[1,10,17,24]

$$H=-\sum_{i.j,\sigma} t_{ij}(c^{\dagger}_{i\sigma}c_{j\sigma}+\text{H.c.}) + \sum_{i,j,\sigma,\sigma'} (ij|1/r|kl)c^{\dagger}_{i\sigma}c^{\dagger}_{j\sigma'}c_{l\sigma'}c_{k\sigma}, \tag{1}$$

where $c^{\dagger}_{i\sigma}$ creates an electron of spin σ in a Wannier orbital at site i, which we denote φ_i. The Coulomb matrix elements are given by the integrals

$$(ij|1/r|kl)=\int d^3r\,d^3r'\,\varphi^*_i(r)\varphi^*_j(r')\frac{e^2}{|r-r'|}\varphi_l(r')\varphi_k(r), \tag{2}$$

and restricting ourselves to only one- and two-center integrals between nearest-neighbors the following matrix elements result:

$$U=(ii|1/r|ii), \tag{3a}$$

$$V=(ij|1/r|ij), \tag{3b}$$

$$J=(ij|1/r|ji), \tag{3c}$$

$$J'=(ii|1/r|jj), \tag{3d}$$

$$\Delta t=(ii|1/r|ij). \tag{3e}$$

Matrix elements involving three and four centers are likely to be substantially smaller than these, as they involve additional overlap factors. Even though the repulsion term (3b) could be of appreciable magnitude for sites farther than nearest neighbors, we assume that such terms will not change the physics qualitatively.

The parameters J and J' describe nearest-neighbor exchange and pair hopping processes, and are always positive by definition. They are in principle equal in magnitude (if the Wannier orbitals are assumed to be real), but we believe it is useful to consider them separately because they give rise to two distinct physical processes, and because their effective magnitude could be different due to wave function renormalization effects. We have found in our previous work that they

both favor ferromagnetism,[13] although the importance of J' is small for large values of the on-site repulsion U. The parameter Δt gives rise to an occupation-dependent hopping rate, which yields a tendency to pairing of carriers at the top of the band.[25]

The Hamiltonian (1), keeping these matrix elements, is of the form

$$H=-\sum_{i,j} t_{ij}^{\sigma}(c_{i\sigma}^{\dagger}c_{j\sigma}+\text{H.c.})+U\sum_{i} n_{i\uparrow}n_{i\downarrow}+V\sum_{\langle i,j\rangle} n_i n_j + J\sum_{\langle i,j\rangle,\sigma,\sigma'} c_{i\sigma}^{\dagger}c_{j\sigma'}^{\dagger}c_{i\sigma'}c_{j\sigma}+J'\sum_{\langle i,j\rangle,\sigma,\sigma'} c_{i\sigma}^{\dagger}c_{i\sigma'}^{\dagger}c_{j\sigma'}c_{j\sigma}, \tag{4a}$$

with

$$t_{ij}^{\sigma}=t-\Delta t(n_{i,-\sigma}+n_{j,-\sigma}). \tag{4b}$$

All matrix elements in Eq. (4) are expected to be always positive, except possibly for the hybrid matrix element Δt. With the convention that the single-particle hopping matrix element t is positive and that the operators $c_{i\sigma}$ describe electrons (rather than holes), the sign of Δt in Eq. (4) is also expected to be positive.[25,26]

A. Mean-field theory

A mean-field decoupling of the interaction terms in Eq. (4) yields for the energies of electrons of spin σ

$$E_{\sigma}(\epsilon)=\left[1-\frac{J}{t}(I_{\uparrow}+I_{\downarrow})-(n-m\sigma)\frac{\Delta t}{t}(I_{\uparrow}+I_{\downarrow})-\frac{J'}{t}I_{-\sigma}+\frac{V}{t}I_{\sigma}\right]\epsilon-\sigma\left[\frac{U+zJ}{2}\right]m+2z\Delta t I_{-\sigma}-\mu, \tag{5}$$

with ϵ the band energy and μ the chemical potential. Here, n is the average occupation per site,

$$n=\langle n_{i\uparrow}+n_{\downarrow}\rangle, \tag{6a}$$

m the average magnetization per site,

$$m=\langle n_{i\uparrow}-n_{\downarrow}\rangle, \tag{6b}$$

and I_{σ} the average bond occupation for spin σ,

$$I_{\sigma}=\langle c_{i\sigma}^{\dagger}c_{j\sigma}\rangle, \tag{6c}$$

with i, j nearest-neighbor sites. If $g(\epsilon)$ is the density of states per site and the band energies ϵ extend from $-D/2$ to $D/2$ (D=bandwidth), we have

$$n=\int_{-D/2}^{D/2} d\epsilon g(\epsilon)[f(E_{\uparrow}(\epsilon))+f(E_{\downarrow}(\epsilon))], \tag{7a}$$

$$m=\int_{-D/2}^{D/2} d\epsilon g(\epsilon)[f(E_{\uparrow}(\epsilon))-f(E_{\downarrow}(\epsilon))], \tag{7b}$$

$$I_{\sigma}=\int_{-D/2}^{D/2} d\epsilon g(\epsilon)\left[\frac{-\epsilon}{D/2}\right]f(E_{\sigma}(\epsilon)), \tag{7c}$$

with f the Fermi function. Equations (7) together with Eq. (5) are solved self-consistently to yield the average magnetization for given value of the electronic density (or chemical potential).

The qualitative effect of all interaction parameters within mean field theory can be inferred from Eq. (5). The bond occupation parameter I_{σ} depends on the band occupation for that spin, n_{σ}. At zero temperature, for a constant density of states,

$$I_{\sigma}(n_{\sigma})=n_{\sigma}(1-n_{\sigma}), \tag{8a}$$

and for a one-dimensional tight-binding band,

$$I_{\sigma}(n_{\sigma})=\frac{1}{\pi}\sin(\pi n_{\sigma}). \tag{8b}$$

In both cases, as well as more generally, the bond occupation is maximum for a 1/2-filled band ($n_{\sigma}=1/2$) and goes to zero as the bottom or top of the band is approached.

In Eq. (5), the interactions U and J give rise to an exchange energy, and J and J' give rise to band narrowing, so that all these parameters favor spin polarization. The nearest-neighbor repulsion V does not affect exchange but only modifies the kinetic energy. Even though it may appear that because it decreases the effective mass (by increasing the coefficient of ϵ) it should disfavor spin polarization, a more detailed analysis of the mean-field equations shows that for low band filling ($n<0.5$) the effect of V is to increase the tendency to spin polarization. The reason is that because of the concave nature of the bond occupation, Eq. (8), as spin polarization develops the cost in kinetic energy of the majority spins is smaller than the gain for the minority spins. The effect is slightly more pronounced for the 1D band than for the flat band because $I_{\sigma}(n_{\sigma})$ is slightly more concave in the former case. This prediction of mean-field theory is not supported by results of exact diagonalization.

The interaction Δt enters both as an exchange contribution and as a modification of the kinetic energy. There is an overall band narrowing effect proportional to $n\Delta t$ which favors spin polarization increasingly as the band filling increases. However, the term in the kinetic energy proportional to $m\Delta t$ *disfavors it* increasingly as the band filling increases. Furthermore, there is an exchange contribution from Δt that is opposite in sign to that of U and J. The overall effect of Δt within mean-field theory is to enhance the tendency to ferromagnetism but progressively *less* so as the band filling increases. This prediction is also in disagreement with results of exact diagonalization, as will be seen in the next section.

The condition on the parameters to give rise to a given magnetization m at zero temperature is obtained by equating the Fermi energies $\epsilon_{F\sigma}$ of up and down electrons. For a bandwidth $D=2zt$ (z = number of nearest-neighbors to a site) and n the band occupation, assuming a flat density of states we have

$$\epsilon_{F\sigma}=\frac{D}{2}[n+\sigma m-1], \tag{9a}$$

$$I_{\sigma}=\frac{1-(1-n-\sigma m)^2}{4}. \tag{9b}$$

Defining the reduced parameters

$$u=\frac{U}{D}, \tag{10a}$$

$$j=\frac{zJ}{D}, \tag{10b}$$

$$j'=\frac{zJ'}{D}, \tag{10c}$$

$$v=\frac{zV}{D}, \tag{10d}$$

$$k=\frac{2z\Delta t}{D}, \tag{10e}$$

Eq. (5) yields for the nearest-neighbor exchange required to give rise to magnetization m

$$j=\frac{1-u-k(2-n)-(j'/2)[1-m^2+(1-n)^2]+(v/2)[1-m^2-3(1-n)^2]}{2-(1-n)^2-m^2}. \tag{11}$$

For a one-dimensional tight-binding band,

$$\epsilon_{F\sigma}=-2t\cos\left[\pi\frac{n+\sigma m}{2}\right], \tag{12a}$$

$$I_\sigma=\frac{1}{\pi}\sin\left[\pi\frac{n+\sigma m}{2}\right]. \tag{12b}$$

In terms of the reduced parameters Eq. (10), the condition equivalent to Eq. (11) is

$$j=c\sin\left(\frac{\pi n}{2}\right)\frac{1-\dfrac{u}{c\sin(\pi n/2)}-k\left[n+\cot\dfrac{\pi n}{2}\left(\dfrac{2}{\pi}+\dfrac{\cos(\pi m/2)}{c}\right)\right]-2j'\dfrac{\cos(\pi m/2)}{\sin(\pi n/2)}-\dfrac{2v}{\pi}\dfrac{\cos(\pi m/2)\cos\pi n}{\sin(\pi n/2)}}{1+(4c/\pi)\sin^2(\pi n/2)\cos(\pi m/2)}, \tag{13a}$$

$$c=\frac{\sin(\pi m/2)}{m}. \tag{13b}$$

These relations display more clearly the effect of the various interaction parameters in mean field theory. For the one-dimensional case it can be seen that as the band filling n approaches 0 (or 2) it becomes easier to obtain ferromagnetism due to the prefactor $\sin(\pi n/2)$. This is because the density of states in one dimension diverges as one approaches the bottom and top of the band. The parameter k (proportional to Δt) is less helpful for ferromagnetism as n approaches the top of the band, as the factor $cot(\pi n/2)$ becomes increasingly negative. The same occurs for the flat band due to the factor $(2-n)$ multiplying k in Eq. (11). The nearest-neighbor repulsion favors or disfavors ferromagnetism depending on whether the sign of $\cos(\pi n)$ is positive or negative, i.e., whether the band is less or more than one-quarter full (for the flat density of states case the dividing point is $n=0.5$ also for full polarization but $n=0.423$ for onset of spin polarization). As we will see in the next section, these predictions are in disagreement with results of exact diagonalization.

B. Exact solutions

Exact solutions of the Hamiltonian (4) are easily obtained for the cases of (1) two electrons in an empty band, (2) two holes in a full band, (3) one overturned spin in the fully polarized half-filled band, and (4) two electrons in a two-site lattice.

1. Two-particles in an empty band

The lowest-energy two-electron state will have zero center-of-mass momentum, assuming $J'>0$. Its wave function is given by

$$|\Psi\rangle=\sum_k f_k c_{k\uparrow}^\dagger c_{k\downarrow}^\dagger|0\rangle. \tag{14}$$

From the Schrödinger equation one finds that the eigenvalue is determined by the equation

$$(u+j')G_0+(v+j)G_2+2kG_1+[k^2-(u+j')(v+j)]\times(G_0G_2-G_1^2)=1, \tag{15a}$$

with

$$G_i=-\frac{1}{N}\sum_k\left(-\frac{\epsilon_k}{D/2}\right)^i\frac{1}{E_0/D-\epsilon_k/(D/2)}, \tag{15b}$$

$$\epsilon_k=-2t\cos k. \tag{15c}$$

All the interaction parameters in Eq. (4) are positive, and this equation does not have any bound-state solutions. For an infinite chain, the ground-state energy is simply

$$E_0=-4t. \tag{16}$$

The ''fully polarized'' wave function for two-particles is of the form

$$|\Psi(p)\rangle = \frac{1}{\sqrt{2}}\sum_k (f_k - f_{p-k}) c^\dagger_{k\uparrow} c^\dagger_{p-k\uparrow}|0\rangle \tag{17}$$

for a triplet pair of center-of-mass momentum p. The eigenvalue equation is

$$\frac{(v-j)}{N}\sum_k \frac{\sin k[\sin k - \sin(p-k)]}{E_0/D - \epsilon_k/D - \epsilon_{p-k}/D} = 1 + \cos p, \tag{18}$$

and the lowest-energy state for a finite chain is obtained for $p = 2\pi/N$. For an infinite chain $p \to 0$ and Eq. (18) becomes

$$\frac{(v-j)}{N}\sum_k \frac{\sin k^2}{E_0/D - \epsilon_k/(D/2)} = 1, \tag{19}$$

which can only yield a lower-energy state than the singlet state, Eq. (18), if

$$J > V. \tag{20}$$

In one spatial dimension, the critical value of J is $J_c = V + 2t$. This ''ferromagnetic state'' is actually a triplet superconducting state, as the particles are bound. For electrons in metals, the condition (20) is likely not to be valid. Hence in this limit singlet and triplet states are degenerate for any values of the interactions assuming Eq. (20) is not satisfied.

2. Two holes in full band

A particle-hole transformation of the Hamiltonian (4) leads to a Hamiltonian of the same form, with hopping amplitude

$$t^\sigma_{ij} = t_h + \Delta t(n_{i,-\sigma} + n_{j,-\sigma}), \tag{21a}$$

$$t_h = t - 2\Delta t, \tag{21b}$$

instead of Eq. (4b). Hence, the eigenvalue equation for two holes with antiparallel spin is of the same form as Eq. (15) *with opposite sign of* Δt, $D = 2zt_h$, and

$$\epsilon_k = -2t_h \cos k, \tag{22}$$

while for two holes of parallel spin the eigenvalue equation is the same as Eq. (19), with ϵ_k given by Eq. (22). Again, assuming $J < V$ the holes will not be bound in a triplet state. However, in this limit a singlet bound-state can exist induced by the interaction Δt. The regime of parameters where such a bound-state occurs is discussed in Ref. 27, for interactions in the Hamiltonian U, V, and Δt; in the presence of J and J' those results still apply with the replacement $U \to U + zJ'$, $V \to V + J$. Thus in this model when the band is almost full ferromagnetism competes with singlet superconductivity.

3. One overturned spin in a fully polarized half-filled band

A particle-hole transformation for spin-up particles leads to a Hamiltonian of essentially the same form as Eq. (4). The Hamiltonian

$$H^{\rm tr} = H^{\rm tr}_0 + U\sum_i n_{i\downarrow} + zJ\sum_i n_{i\uparrow}, \tag{23}$$

with

$$H^{\rm tr}_0(U,V,J,J') = H(-U,-V,J',J), \tag{24}$$

describes both the states with one overturned spin (one spin-down particle and one spin-up hole) and the fully polarized state. Equation (4b) for the hopping amplitudes is replaced by

$$t^\sigma_{ij} = t - \sigma\Delta t(n_{i,-\sigma} + n_{j,-\sigma} - 1 + \sigma). \tag{25}$$

The fully polarized ferromagnetic state corresponds to the vacuum of $H^{\rm tr}$ and has energy $E = 0$. States with one overturned spin correspond to states of $H^{\rm tr}$ with two-particles of opposite spin, and the lowest energy is

$$E = E_0 + U + zJ, \tag{26}$$

where E_0 is the ground-state eigenvalue of $H^{\rm tr}_0$, satisfying the equation

$$(j-u)G_0 - (v-j')G_2 + (u+j)(v-j')(G_0G_2 - G_1^2) = 1, \tag{27}$$

and G_i is given by Eq. (15), with t replaced by $t - \Delta t$ and $D = 2z(t - \Delta t)$. Note that in contrast to Eq. (15), Δt does not enter explicitly into Eq. (27), because it enters with opposite signs for electrons and holes and thus cancels out. Now the on-site interaction as well as the nearest-neighbor interaction between the two-particles (i.e., the up hole and down electron in the original model) is attractive, so that bound states can exist. The condition for the ferromagnetic state to be stable under one spin flip is

$$E_0 + U + zJ < 0, \tag{28}$$

which, in the parameter regime where there is no bound state, yields

$$J > 2t - \frac{U}{z}, \tag{29}$$

which is the same as the Hartree-Fock solution, Eq. (11) or Eq. (13), for $m = n = 1$. The fact that the up hole and down electron are unbound implies that the system is metallic. For sufficiently large U and V, however, a bound state exists and the condition for ferromagnetism is more stringent than Eq. (29).

The quantities G_1 and G_2 can be written in terms of G_0 as

$$G_1 = 1 - aG_0, \tag{30a}$$

$$G_2 = a^2G_0 - a, \tag{30b}$$

$$a = \frac{E_0}{D}. \tag{30c}$$

In one spatial dimension,

$$G_0 = -\frac{1}{\sqrt{a^2 - 1}}, \tag{31}$$

and the condition for ferromagnetism, in the regime where a bound state exists, is found to be

$$4j\{u-(v-j')[1+j(v-j')-2j(u+j)]\}>1. \quad (32)$$

In higher dimensions the integral for G_0 can be written in terms of the Watson integral.[27] In the limit of large U, the condition on ferromagnetism resulting from these equations is

$$J>\frac{2t^2}{U+J'-V}, \quad (33)$$

which is the result from lowest-order strong-coupling perturbation theory. The next-order correction can be worked out following the steps in Ref. 16.

4. Two electrons in a two-site lattice

The ground-state energy for two electrons in a two-site chain is easily obtained analytically (see also Ref. 24). The energies of the lowest singlet and triplet states are

$$E_S=\frac{U+J'+V+J}{2}-\sqrt{\left(\frac{U+J'-V-J}{2}\right)^2+4(t-\Delta t)^2}, \quad (34a)$$

$$E_t=V-J, \quad (34b)$$

respectively, so that in the absence of J the singlet energy is always lower than the triplet. The condition for ''ferromagnetism,'' $E_t<E_s$, in this ''half-filled band'' system yields

$$J>\sqrt{\left(\frac{U+J'-V}{2}\right)^2+2(t-\Delta t)^2}-\frac{U+J'-V}{2} \quad (35)$$

or, for the case $J'=J$,

$$J>\sqrt{\left(\frac{U-V}{2}\right)^2+(t-\Delta t)^2}-\frac{U-V}{4}. \quad (36)$$

For large U, Eq. (35) reduces to Eq. (33). More generally, these relations show that in the dimer U, Δt, and J' help ferromagnetism while V opposes it. These qualitative features are found to persist in larger systems and for other band fillings.

III. NUMERICAL RESULTS

We obtain the ground-state energy of the Hamiltonian (4) for one-dimensional chains in each total spin sector by exact diagonalization of the Hamiltonian. Most of the numerical results were obtained for chains of $N=8$ sites, for which we can study band fillings $n=N_e/N=0.25,0.5,\ldots,1.75$. We also obtained some results for other band fillings by diagonalization of chains of 6, 10, and 12 sites, which were found generally to smoothly interpolate between the results for band fillings of the 8-site chain.

A. Effect of band filling

We start by examining the band filling dependence for the Hamiltonian with interactions U and J only. For the half-filled band case, we had found that mean-field theory gave results in reasonable agreement with exact diagonalization.[15,13]

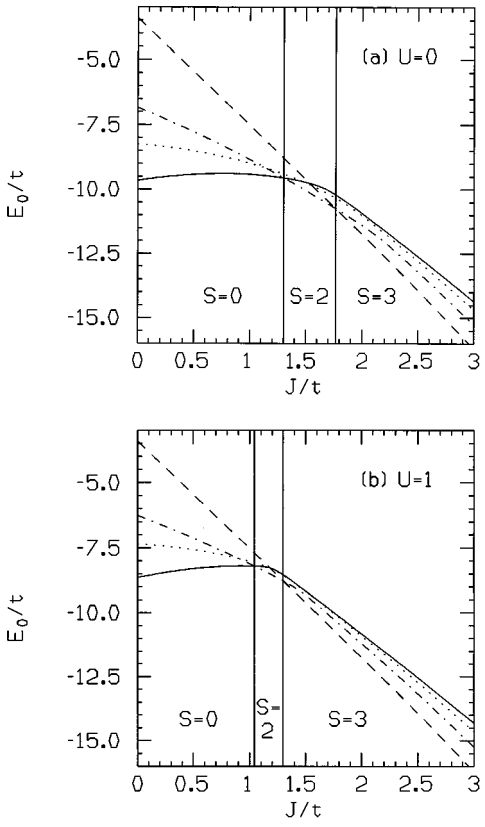

FIG. 1. Ground-state energy vs J for band filling $n=0.75$ ($N=8$, $N_e=6$) for (a) $U=0$ and (b) $U=1$. Here and in the following figures, $t=1$ and other parameters in the Hamiltonian (4) are zero unless otherwise indicated. The solid, dotted, dash-dotted, and dashed lines correspond to lowest-energy states with total spin $S=0$, $S=1$, $S=2$, and $S=3$, respectively. As J increases, the lowest-energy state has subsequently $S=0$, $S=2$, and $S=3$ (full polarization). The regime of partial polarization ($S=2$) becomes smaller as U increases.

Figure 1 shows the behavior of ground-state energy versus J for two values of U, for band filling $n=0.75$. Similarly, as for the half-filled band case, a regime of partial spin polarization exists for small U, which becomes smaller as U increases and disappears for large U. Similar behavior is found for other band fillings. However, it is difficult to extract reliable results for partial spin polarization regimes from small chains because of the importance of finite-size effects. In the following we concentrate on the conditions necessary for full spin polarization.

Figure 2 shows the value of J needed for full spin polarization as function of on-site repulsion for various band fillings. As in the half-filled band case, increasing U is favorable to spin polarization. However, for $n<1$ the value of J needed for ferromagnetism remains finite as $U\to\infty$ and increases as n decreases. This is qualitatively different from the predictions of mean-field theory shown in Fig. 2(b), where it generally becomes easier to spin polarize as the band filling decreases. Similarly, in Fig. 3 we plot the phase boundaries for full polarization in the J-n plane for various

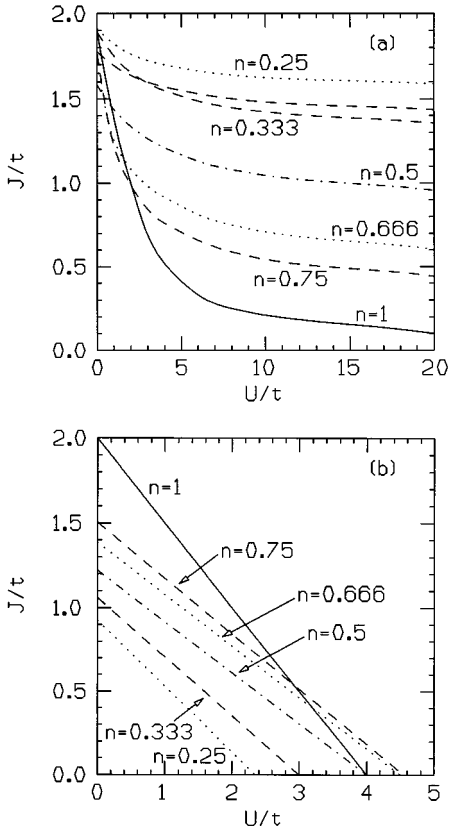

FIG. 2. Phase boundaries for full spin polarization in the U-J plane for different band fillings. (a) Exact-diagonalization, (b) mean-field theory. In (a) the results for $n=0.666$ were obtained from a 6-site chain and those for $n=0.333$ from both a 6-site chain (lower line) and a 12-site chain (upper line); all other boundaries were obtained from an 8-site chain. Above each line, the ground-state is fully spin polarized for that band filling.

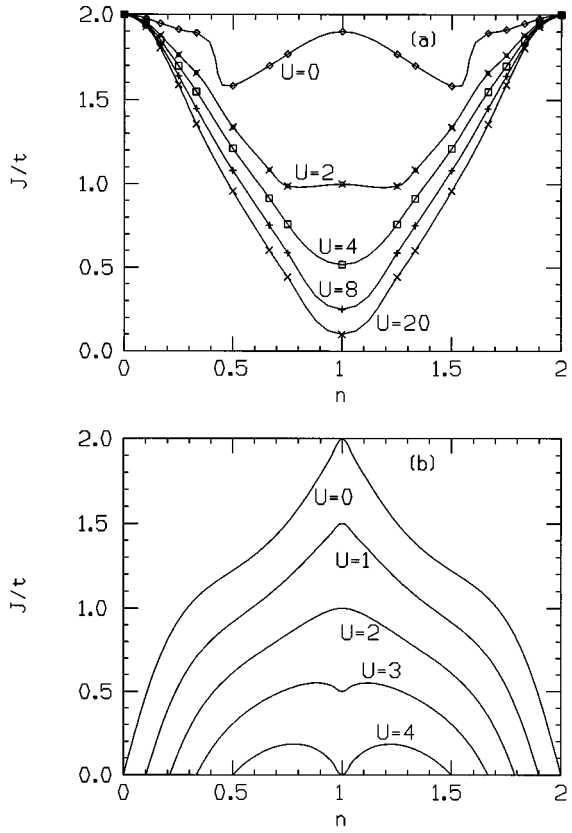

FIG. 3. Phase boundaries for full spin polarization as a function of J and band occupation n for various values of U. (a) Exact diagonalization, (b) mean-field theory. In (a) numerical results are indicated by symbols and the lines are smoothly drawn through the data to guide the eye. Full spin polarization occurs above each line for the corresponding value of U. Results for $n\leqslant 0.25$ were obtained from the exact solution for two particles in chains of increasing size.

values of U. While the qualitative effect that U makes spin polarization easier is reproduced by mean-field theory, the band filling dependence predicted by mean-field theory is opposite to that found in the exact solution.

Mean-field theory clearly fails because electrons with antiparallel spin will correlate their motion to avoid both the on-site repulsion U and the bond-charge repulsion J, thus making it less favorable to spin polarize than an uncorrelated (mean-field) wave function would predict. In the half-filled band case such a correlation is not possible because there is no room for electrons to avoid each other both on sites and on bonds; this is why mean-field theory in the presence of U *and* J works reasonably well. As the band filling decreases (or increases) from one-half, there is increasing room for electrons (or holes) to avoid each other, and mean-field theory becomes increasingly inaccurate. The discrepancy between exact and mean-field results is enhanced by the fact that the density of states diverges as the band edge is approached in the one-dimensional model. For a flat density of states, mean-field theory in fact predicts a decrease in the tendency to ferromagnetism as one moves away from half-filling.

B. Effect of nearest-neighbor repulsion

A realistic model of electrons in metals, if it includes the nearest-neighbor exchange J, should also include the nearest-neighbor repulsion V which will in general be larger in magnitude than J. In fact in the absence of V the model Hamiltonian (4) will also be unstable towards triplet superconductivity, as electrons of parallel spins attract each other. Since the interaction between electrons of parallel spin is $(V-J)$, that instability will disappear for $V\geqslant J$ which is expected to be the case for electrons in metals.

Numerical results show that V suppresses the tendency to ferromagnetism, particularly for small U, for all band fillings. Figures 4 and 5 show the effect of V for band fillings $n=1$ and $n=0.5$. As discussed in Sec. II, mean-field theory predicts no effect of the interaction V on the condition for full spin polarization at these band fillings.

In Fig. 5(a), we show also the results of the exact calculation of the boundary of stability of the fully ferromagnetic state to a single spin flip [Eq. (28)]. For small nearest-neighbor repulsion it agrees with the exact diagonalization, while for large V it yields a lower value of J. The reason is that in the latter regime the fully polarized state becomes unstable to more than one spin flip, in fact to the state with $S=0$. This is easily understood for very large V, where the fully polarized state becomes unstable towards a charge-density-wave state with $S=0$ well before it would become unstable to flipping a single spin as J decreases. Even for

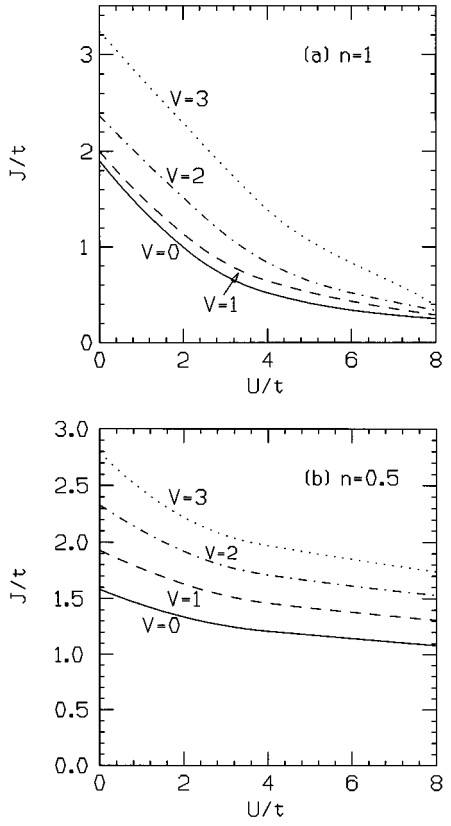

FIG. 4. Phase boundaries for full spin polarization for band fillings (a) $n=1$ and (b) $n=0.5$ from exact-diagonalization for various values of the nearest neighbor repulsion V.

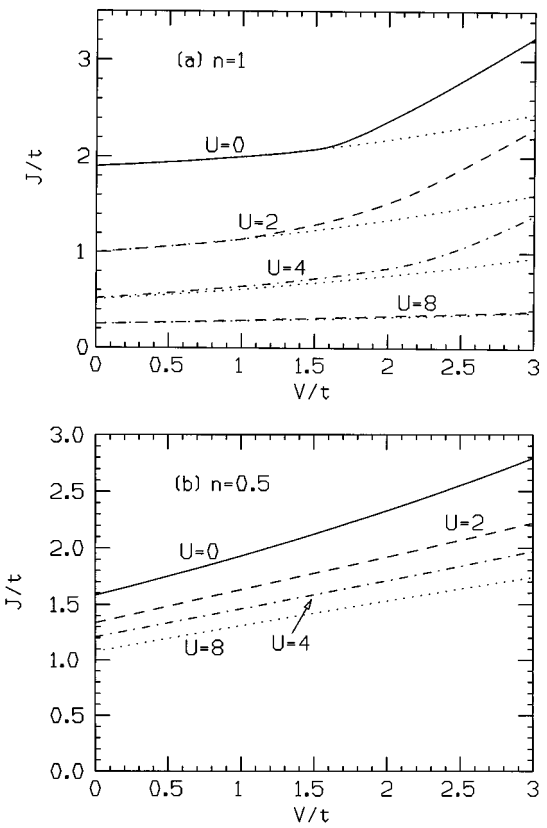

FIG. 5. Same as Fig. 4, with the phase boundaries plotted vs V for various values of U. The dotted lines in (a) indicate the exact boundaries of stability of the fully polarized state with respect to the state with one spin flipped, Eq. (28). Note that they coincide with the exact diagonalization results only for small U and V. The difference is small for large U.

$V=0$, the ferromagnetic state becomes unstable to states with more than one spin flip as U increases, and for $U=4$ and $U=8$ in Fig. 5(a) the exact-diagonalization results are slightly larger than the analytic boundary of stability for all values of V.

To obtain an estimate of the effects of finite-size in this calculation we examined the size dependence of the analytic boundary results. Even for $N=8$ the results differ from the infinite chain limit by less than 5% for all interaction values. Finite-size effects are largest for $U=V=0$ and decrease rapidly as the interactions increase. These results are shown in Fig. 6. Note that in the infinite system the phase boundary is independent of V for small U and V, so that the dependence found for $N=8$ in that regime is a finite-size effect.

It can be seen that for small U no ferromagnetism exists unless J is substantially larger than V, which is unphysical. To obtain a better appreciation of the physical parameter range where ferromagnetism occurs we plot in Fig. 7(a) the value of J required for full spin polarization assuming $V=2J$. No ferromagnetism is obtained for small U under this assumption. For intermediate U, if J (and V) become too large, ferromagnetism also disappears [dashed line boundaries in Fig. 7(a)]. We also show mean-field results for comparison [Fig. 7(b)], which agree qualitatively only in showing that as n decreases from 1 initially larger values of J are required for ferromagnetism. Figure 8 shows the phase boundaries in the J-n plane under the condition $V=2J$ from exact and mean-field solutions. Comparing the exact results with Fig. 3(a) it is seen that in the presence of nearest-neighbor repulsion ferromagnetism is strongly suppressed for small U and low (or high) values of band filling.

C. Effect of pair hopping

The effect of the pair hopping interaction J' on ferromagnetism was considered in the half-filled band case in Ref. 13,

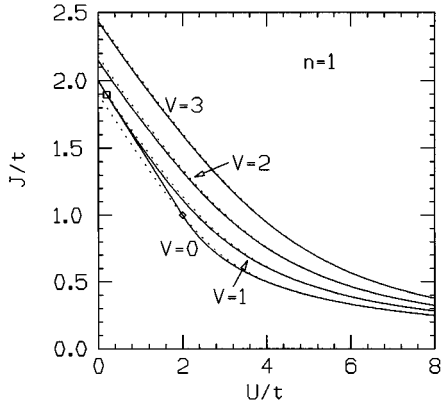

FIG. 6. Comparison of results for 8-site chain (dotted lines) and infinite chain (solid lines) from the analytic criterion for stability of the fully polarized state in the half-filled band, Eq. (28). The open square and diamond symbols indicate the points where a bound state develops for $V=1$ and $V=0$, respectively.

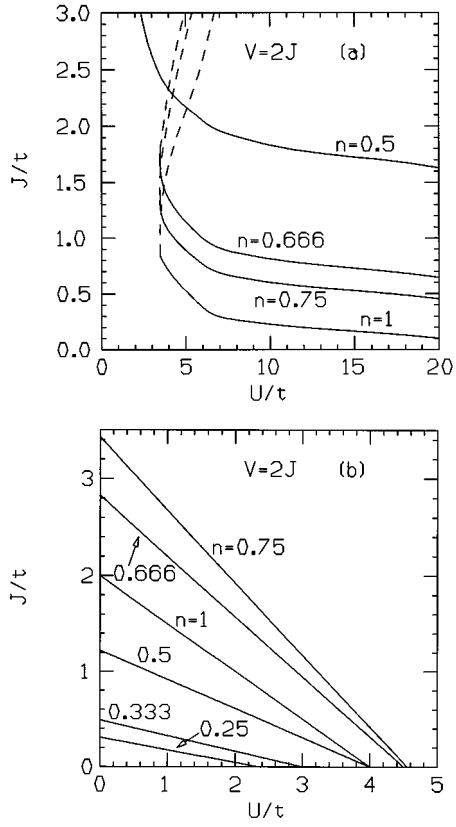

FIG. 7. Phase boundaries for full spin polarization for $V=2J$. (a) Exact diagonalization, (b) mean-field theory. The dashed lines in (a) indicate the boundaries of stability of the fully polarized state for large J and V. For J (and V) larger than given by those boundaries the unpolarized state is again the ground-state.

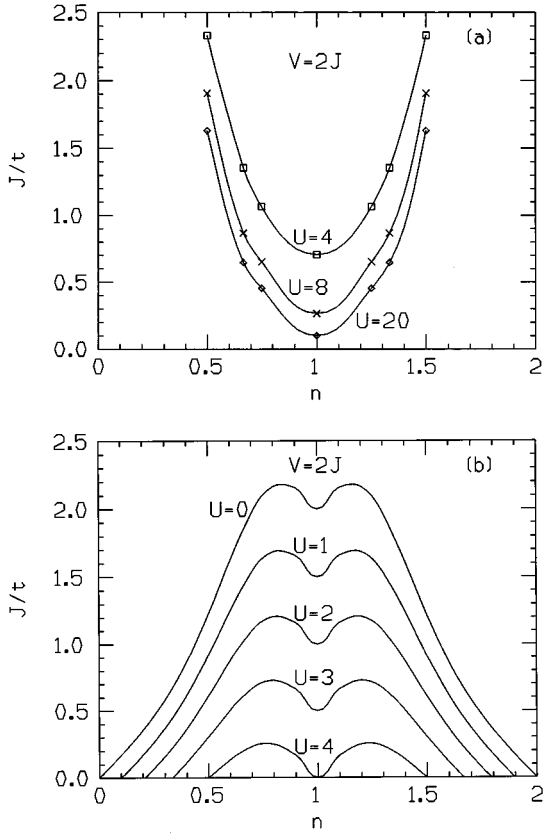

FIG. 8. Phase boundaries for full spin polarization for $V=2J$ in the J-n plane. (a) Exact diagonalization, (b) mean-field theory. In the exact solution, ferromagnetism is strongly suppressed for small U compared to the case $V=0$.

and is found to be similar for other band fillings. While J' by itself will not give rise to ferromagnetism, it enhances the tendency to ferromagnetism in the presence of J. This is shown in Fig. 9, which displays the boundaries to full spin polarization for $J'=0$ and $J'=J$ for various band fillings. The effect of J' becomes small for large U as double-site occupancy is suppressed.

The qualitative fact that J' enhances the tendency to ferromagnetism is correctly reproduced by mean-field theory, although it greatly overestimates the effect of J' except for half-filling, where mean-field theory predicts no dependence on J' [Fig. 9(b)]. The exact phase boundary calculation for $n=1$ shows that for $U<2t$ the phase boundary is independent of J' for an infinite chain [Eq. (29)] but depends somewhat on J' for finite N, in agreement with Fig. 9(a). That is, the difference in the results for $J=0$ and $J=J'$ in Fig. 9(a) for small J and $n=1$ is a finite-size effect. For $U>2t$ there is an effect of J' in the exact solution even for an infinite chain, given accurately by the results in Fig. 9(a) except for U very close to $2t$. These exact results for an infinite chain are shown in Fig. 9(c).

Thus, for a half-filled band J' has no effect on the condition for full spin polarization both for small U and in the limit of large U, and has a small effect for intermediate U. As seen in Ref. 13 it also has an appreciable effect on the condition for *onset* of spin polarization, particularly for small U. For other band fillings, the results in Fig. 9(a) suggest that J' has an effect on the condition for full polarization also for small U. As $n\rightarrow 0$, however, the critical value of J, obtained from Eq. (19), is again independent of J' in the infinite system limit.

D. Effect of hybrid interaction

Next we consider the effect of the hybrid interaction Δt. Δt breaks electron-hole symmetry and gives rise to increasing band narrowing as the band filling increases, since from Eq. (4b) the effective hopping is

$$t_{\mathrm{eff}}(n)=t-n\Delta t, \tag{37}$$

which suggests that ferromagnetism should become easier as n increases due to the increasing density of states. However, as seen in Sec. II, mean-field theory predicts the opposite behavior when the effect of Δt on the exchange energy is taken into account, both for the flat band and the one-dimensional case. This is of concern because it would suggest that ferromagnetism should occur preferentially for bands with electronlike cariers, while in nature it is seen to occur preferentially for holelike carriers.

Results of exact-diagonalization are shown in Fig. 10, where we compare the effect of Δt when the band is less than half-full ($n=0.5$) and more than half-full ($n=1.5$). We also show the results obtained in the absence of Δt with an effective hopping given by Eq. (37). The effect of Δt is seen

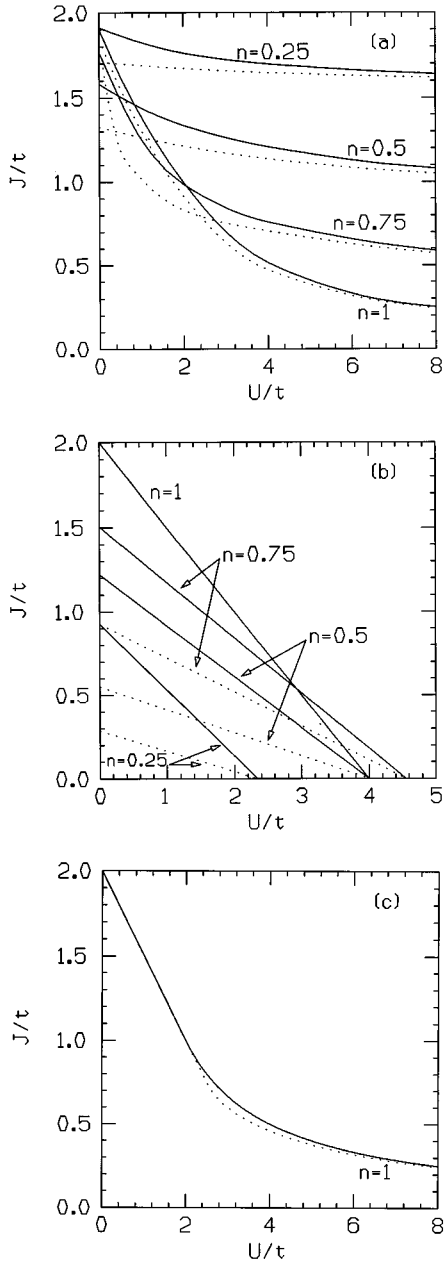

FIG. 9. Phase boundaries for full spin polarization in the presence of pair hopping: $J'=J$ (dotted lines) compared with $J'=0$ (solid lines). (a) Exact diagonalization (8-site chain), (b) mean-field theory, and (c) exact phase boundary for $n=1$ [Eq. (28)], infinite chain.

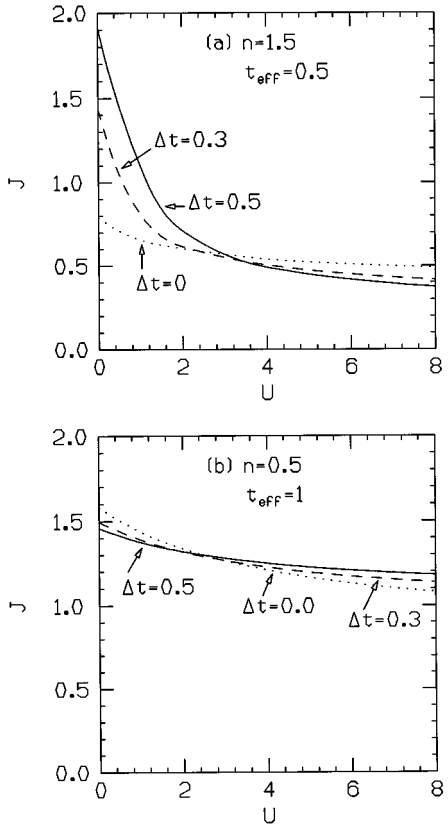

FIG. 10. Phase boundaries for full spin polarization in the presence of the hybrid interaction Δt for (a) holes, band more than 1/2-filled and (b) electrons, band less than half-filled. In (a), the values of t used are $t=0.95$ and $t=1.25$ for $\Delta t=0.3$ and $\Delta t=0.5$, respectively, so that $t_{\rm eff}=0.5$ for both values of Δt at $n=1.5$ [see Eq. (37)]. In (b), the values of t used are $t=1.15$ and $t=1.25$ for $\Delta t=0.3$ and $\Delta t=0.5$, respectively, so that $t_{\rm eff}=1$ for both values of Δt at $n=0.5$. Results with $\Delta t=0$ and $t=t_{\rm eff}$ are also shown in both cases (dotted lines).

to be large (small) for more (less) than half-filled bands. Interestingly, for small U the effect of Δt is seen to be in qualitative agreement with mean-field theory: It helps ferromagnetism for $n<1$ and supresses it for $n>1$. However, as U increases it is seen that the effect of Δt reverses, and in fact for the band more than half-full Δt helps ferromagnetism even beyond the band narrowing effect given by Eq. (37). Figure 11 shows the phase diagram in the J-n plane in the presence of Δt compared with mean-field theory. It can be seen that generally Δt gives rise to strong preference to ferromagnetism in the regime above half-filling compared to below half-filling. Mean-field theory [Fig. 11(b)] predicts the opposite behavior.

Finally, Fig. 12 shows the phase boundaries to full spin polarization for a case where all interaction parameters in the Hamiltonian (4) are included, with $V=2J$ and $J'=J$. In the presence of all short-ranged interactions likely to exist in a real system, ferromagnetism is seen to occur for not too large values of the nearest-neighbor exchange interaction for band fillings around $n=1$ and larger.

IV. CONCLUSIONS

We have studied the effect of band filling and of various interaction parameters on the tendency to ferromagnetism in a single-band tight-binding model. The purpose of this study was to shed light on the question whether such a model may approximately describe metallic ferromagnetism in real materials, and in particular whether the nearest-neighbor exchange interaction J plays the dominant role. Another goal of this study was to assess the validity of mean-field theory to describe the properties of this Hamiltonian.

In previous work we had considered the Hamiltonian with interaction parameters U and J. It was found that within mean-field theory this Hamiltonian generically gives rise to ferromagnetism with partial spin polarization independent of

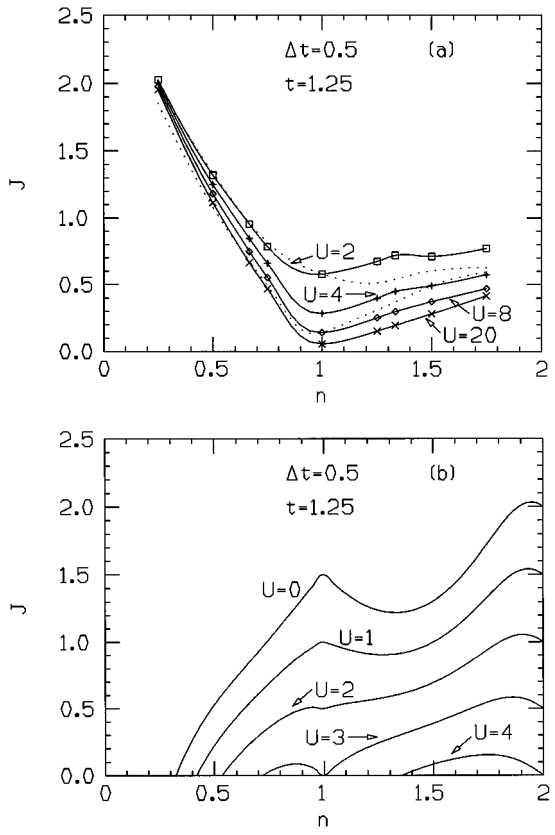

FIG. 11. Phase boundaries for full spin polarization in the J-n plane in the presence of hybrid interaction $\Delta t=0.5$, with $t=1.25$. (a) Exact diagonalization, (b) mean-field theory. In (a), dotted lines show the phase boundary obtained for $U=2$ (upper) and $U=8$ (lower) with t_{eff} given by Eq. (37).

details of the density of states, which does not happen in the absence of the parameter J (Stoner model). Furthermore, mean-field theory predicted that as U increases ferromagnetism could occur for arbitrarily small values of J for any band filling. An exact diagonalization study for the one-dimensional half-filled band confirmed these predictions qualitatively: A regime of partial spin polarization was found to exist for small U, and the critical J to give rise to ferromagnetism approaches 0 as $U\rightarrow\infty$. This last result was found to be applicable more generally to the half-filled band case in any dimension.[16] Furthermore, in the half-filled band we had found that the pair hopping interaction J' also helps ferromagnetism,[13] in qualitative agreement with mean-field theory. This can be understood since J' and J together give rise to ''bond-charge repulsion,''[20] which is reduced when spin polarization develops.

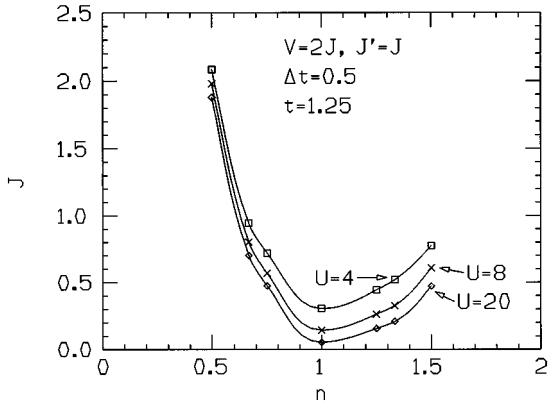

FIG. 12. Phase boundaries for full spin polarization in the J-n plane for a case where all parameters in the Hamiltonian (4) are nonzero, as indicated in the figure.

In this work we studied other band fillings by exact-diagonalization, as well as the effect of other Coulomb interaction parameters, the nearest-neighbor repulsion V, and the hybrid interaction Δt. For band fillings other than $n=1$ it was found that ferromagnetism is less easily achieved within this model, and in particular even for $U\rightarrow\infty$ finite values of J are required for ferromagnetism. We believe this result is likely to hold in higher dimensions also. While mean-field theory predicts the opposite behavior in 1D (that is, that the tendency to ferromagnetism increases away from half-filling), it would predict qualitatively the same trend for any density of states that does not increase rapidly as one approaches the band edges as the one-dimensional one does, in particular a flat density of states or a density of states arising from nearest-neighbor tight-binding models. It is not surprising that predictions of mean-field theory that are sensitive to details of the noninteracting density of states are not reliable.

The effect of the nearest-neighbor repulsion V was found to be to disfavor ferromagnetism, particularly for small U, for all band fillings. In particular, for the case $V=2J$ no ferromagnetism was found for small values of U. However, in real materials where ferromagnetism occurs the on-site repulsion U is likely to be of appreciable magnitude, in which case the effect of V is much less significant. V has also the important effect of suppressing triplet superconductivity, which would occur in this model for $V<J$, a parameter range not likely to apply to electrons in metals. The competition between ferromagnetism and triplet superconductivity predicted by this model, however, could be of relevance to other real systems such as ^{3}He and will be considered elsewhere.

The pair hopping interaction J' was found to enhance the tendency to ferromagnetism for all band fillings, although the effect is quantitatively small especially for large U. Mean-field theory predicts qualitatively the same effect. The qualitative effects of V and J' found here for all band fillings are also consistent with the results found by Strack and Vollhardt[22] and Campbell *et al.*[24] in the half-filled band case.

The hybrid interaction Δt was found to enhance the tendency to ferromagnetism of electronlike versus holelike carriers for small U, and to have the opposite effect for large U. In particular, in the latter case, which is likely to apply to real materials, the enhancement produced by Δt is even larger than the one expected due to the average band narrowing produced by Δt as the band filling increases. Thus for U larger than the bandwidth, as expected for real transition metals, the model strongly favors ferromagnetism for holelike versus electronlike carriers, as observed in nature. This finding from exact diagonalization is especially interesting as it is opposite to the prediction of mean-field theory. We believe that the qualitative effects of the hybrid interaction and nearest-neighbor repulsion found in the one-dimensional model are likely to be similar in higher dimensions.

In general, mean-field theory was found to yield qualitatively incorrect predictions for the effects of the interactions V and Δt. Thus it is not very useful as an analytic method to describe the physics of this generalized tight-binding model. On the other hand, the exact diagonalization study showed that these interactions do not qualitatively change the physics of the model with interactions U and J only as far as ferromagnetism is concerned. Quantitatively, the effect of V is small for large U, and the main effect of Δt can be taken into account by an effective hopping $t_{\rm eff}=t-n\Delta t$. Also the parameter J' was found not to change the physics significantly. Furthermore, no combination of parameters in the Hamiltonian that did not include the nearest-neighbor exchange J was found to be able to induce ferromagnetism. Thus, we argue that the simple model with interactions U and J and hopping $t_{\rm eff}$, with the magnitude of these parameters renormalized by the presence of other interactions, is a useful paradigm to describe the physics of ferromagnetism in metals. For this simple ''U-J model,'' mean-field theory provides a fairly reliable description, more accurate than what the ''Stoner model'' does for the simple Hubbard model and with considerably richer features.[12–17]

Concerning the applicability of this model to real materials, more work is needed to estimate the magnitude of these parameters in particular cases, and examine possible effects of higher dimensionality in exact diagonalization. Still, the present study shows that for reasonable parameter values in the model ferromagnetism is predicted for more than half-filled bands. According to our interpretation of the Slater-Pauling diagram for transition metals[12] Fe would correspond in the effective single-band description to band filling close to 1/2, and Co and Ni to progressively higher fillings. For example, the width of the t_{2g} band in Fe is approximately 5.4 eV.[28] For a nearest-neighbor tight-binding band in a bcc lattice the bandwidth is $D=16t_{\rm eff}$; hence in Fig. 12 a parameter of magnitude 1 corresponds to 0.45 eV (since the effective hopping at half filling in Fig. 12 is 0.75). It can be seen that the required values of J are not unreasonable.

In summary, we have seen that the key parameter in the model is the nearest-neighbor exchange interaction J and that a large value of the on-site repulsion U is important only insofar as it allows the nearest-neighbor exchange to be effective even if it is small in magnitude. In contrast to other models that favor ferromagnetism for a low density of carriers, this model predicts that ferromagnetism is more likely to occur close to a half-filled band, and the hybrid interaction Δt allows the region where ferromagnetism is favored to extend to higher than half-filled band cases. Thus the predictions of this model are consistent with the fact that ferromagnetism is found in nature in the elements Fe, Co, and Ni but not in transition metals at the beginning of the $3d$ series.

ACKNOWLEDGMENTS

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