Ground-state ferromagnetism in a doubly orbitally degenerate model

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In the present paper the ground state of a doubly orbitally degenerate model at weak intra-atomic interaction is studied using the Green function method. Besides the diagonal matrix elements of electron-electron interactions the model includes correlated hopping integrals and interatomic exchange interaction. The influence of orbital degeneracy with Hund's rule coupling, correlated hopping and interatomic direct exchange on the ferromagnetic ordering is investigated. The expressions for ground-state energy and magnetization, the criterion of transition from paramagnetic to ferromagnetic ground state as functions of the model parameters are obtained. The obtained results are compared with some experimental data for magnetic materials.

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I. INTRODUCTION

The problem of an origin of the metallic ferromagnetism, in spite of the variety of theoretical attempts to solve it, still remains open. Nowadays we can distinguish a few ways to obtain the ferromagnetic solution. First, one can consider the Hubbard model[1–3] that describes itinerant electrons in a single nondegenerate band interacting via on-site Coulomb repulsion U. Within this model the ferromagnetic solution has been obtained by means of some approximations,[4,5] also the exact result of Nagaoka[6] shows that the ground state of a band with exactly one electron above or below half-filling is ferromagnetic at $U=\infty$. However, in spite of the large number of papers (for recent reviews see Refs. 4 and 7) the question of an existence of ferromagnetic ordering in the Hubbard model is still under discussion. Second, one can include in a model Hamiltonian, in addition to the intra-atomic Coulomb repulsion, also other matrix elements of electron-electron interaction, which can provide new mechanisms of ferromagnetism stabilization. Third, one can take into consideration the orbital degeneracy with the intra-atomic Hund's rule exchange interaction that forms atomic magnetic moments. Taking into consideration the orbital degeneracy is essentially important: in the case of strong intra-atomic interactions the kinetic superexchange has a ferromagnetic nature, in contrast with the single-band Hubbard model where it has an antiferromagnetic one. We believe that at least for understanding of the ferromagnetism origin in crystals with narrow energy bands it is enough to consider other matrix elements of electron correlations (in addition to the intra-atomic Coulomb repulsion) and orbital degeneracy with intra-atomic Hund's rule exchange.

The importance of orbital degeneracy and Hund's rule exchange interaction for ferromagnetism was first suggested by Slater[8] and van Vleck:[9] the presence of orbital degeneracy and Hund's rule exchange interaction leads to so-called ''atomic ferromagnetism''; in such a situation translational motion of electrons forces the spins of electrons on nearest-neighbor atoms to align in parallel. This mechanism of ferromagnetism based on a microscopic model was studied by Roth[10] considering the Hubbard model with double orbital degeneration. In Refs. 11–14 it was found that ferromagnetism in such two-band Hubbard model coexists with orbital ordering for the case of a quarter-filled band and strong intra-atomic interactions. Moreover, recently Spałek with co-worker[15] have proposed the mechanism of a coexisting ferromagnetism and spin-triplet paired state in a doubly orbitally degenerate band due to the intra-atomic Hund's rule coupling. The problem of metallic ferromagnetism in the two-band Hubbard model has attracted much attention of researchers in a series of papers by means of the dynamical mean-field theory,[16–18] the slave-boson method[15,19] and the Gutzwiller variational wave-function approximation.[20–23] They all find ferromagnetism to be stabilized by intra-atomic Hund's rule coupling at intermediate and strong intra-atomic Coulomb interactions. On the other hand, the authors of Refs. 24 and 25 have obtained that intraatomic Hund's rule exchange does not play the important role in ferromagnetic ordering of crystals with narrow energy bands, and even may destabilize ferromagnetism. In particular, Hirsch[24] suggests that orbital degeneracy and intra-atomic exchange interaction are not likely to play a significant role in the ferromagnetic ordering of Ni or Ni-Cu and Ni-Zn alloys, and a single-band model with interatomic direct exchange contains the essential physics of metallic ferromagnetism of transition-metal compounds. Nolting and co-workers[25] have obtained that magnetization of the doubly orbitally degenerate Hubbard model strongly decreases with increasing intra-atomic Hund's rule exchange for a wide range of model parameters, namely, the intra-atomic exchange coupling substantially suppresses ferromagnetic order.

In the papers cited above the authors have not taken into consideration so-called ''off-diagonal'' matrix elements of electron-electron interaction that are one of the possible mechanisms of ferromagnetism in narrow energy bands as mentioned above. The importance of these matrix elements in a single-band case was pointed out in many works.[4,16,26–35] Here we note the special role of direct exchange interaction and correlated hopping (taking into account of the interatomic density-density Coulomb interaction that plays the essential role in charge ordering goes beyond the goal of this article). In the last 10 years the problem of importance of interatomic exchange interaction for the metallic ferromagnetism again is under discussion in a number of works,[4,16,26–34] where it was concluded that the interatomic exchange interaction J plays a fundamental role for

the stabilization of ferromagnetic ordering in a single-band model. The authors found that the interatomic direct exchange $J>0$ stabilizes ferromagnetic ordering in the single-band Hubbard model at intermediate to strong intra-atomic Coulomb interactions.[4,16,31,34] In the case of weak interactions the interatomic exchange is important for the stabilization of incomplete ferromagnetism.[27,30] Hirsch[30] argued that the interatomic exchange interaction is the main driving force for metallic ferromagnetism in systems like iron, cobalt, and nickel. Also for the special cases of generalized Hubbard models it has been found[32,33] by means of exact techniques that interatomic exchange plays a dominant role in the occurrence of ferromagnetism.

The importance of correlated hopping for understanding of the metallic ferromagnetism in narrow energy band was discussed in Refs. 16,26–29,31–33 and 35. In particular, a generalization of Nagaoka's theorem has been proved,[33] and it has been shown[36,31] that in strong-coupling regime close to half-filling correlated hopping favors ferromagnetism stronger for electronlike carriers than for holelike carriers (the reverse situation occurs at weak interactions[31]). Note, that due to an additional mechanism of correlated hopping[27] at weak intra-atomic interactions the situation, which is analogous to that of strong interactions, can be realized: correlated hopping favors ferromagnetism stronger for electronlike carriers versus holelike carriers (see also Sec. III).

In this connection, the necessity of a further study of the metallic ferromagnetism problem in narrow energy bands is obvious. First, it is interesting and important to find how the orbital degeneracy with intra-atomic Hund's rule coupling and ''off-diagonal'' matrix elements of electron-electron interaction (correlated hopping and inter-atomic direct exchange interaction) in the aggregate show itself. Note that these studies were performed partially in Ref. 24 by means of exact diagonalization, in particular, for the case of small one-dimensional chains and strong intra-atomic Coulomb repulsion the role of orbital degeneracy with intra-atomic Hund's rule coupling and interatomic exchange interaction for the ferromagnetic ordering was studied. However, the results depend sensitively on the number of lattice sites and the boundary conditions, on the one hand, and the study is restricted to the one-dimensional case, on the other hand. Second, there is a contradiction about the role of intra-atomic Hund's rule interaction for the stabilization of ferromagnetic ordering, as mentioned above. Therefore, the present paper is devoted to the study of the metallic ferromagnetism problem.

Our consideration of the metallic ferromagnetism problem is the development of the Stoner theory following the ideas proposed in Refs. 24, 30, and 31 on the basis of more complete model Hamiltonian[37] than the Hamiltonians that have been used previously. We assume (similar to the Refs. 24 and 30, and 31) that the mean-field approximation can give the qualitatively correct physical picture in the case of the intermediate electron-electron correlations. This approach leads to the results that agree qualitatively with those of the Refs. 4,15,17,23–25 and 38 where the authors use other methods to treat electron correlations, and are in agreement with experimental data. In particular, the approach used by us allows to avoid the problem of Curie temperature overestimation (in this connection see Ref. 39), which is typical for the usual mean-field theories (of Stoner type).

The structure of the paper is the following. In Sec. II we formulate the Hamiltonian of the doubly orbitally degenerate Hubbard model that is generalized by taking into account correlated hopping and interatomic exchange interaction. The single-particle Green function and energy spectrum at arbitrary values of electron concentration are derived by means of the mean-field approximation. In Sec. III the ferromagnetism in ground state of the model is investigated. The role of orbital degeneracy with intra-atomic Hund's rule coupling, of correlated hopping, and of interatomic direct-exchange interaction for the stability of ferromagnetic ordering is studied. The expressions for ground-state energy and magnetization as functions of the model parameters, the criterion of transition from paramagnetic to ferromagnetic ground state are found. Finally, Sec. IV is devoted to the conclusions from the obtained results.

II. GREEN FUNCTION AND ENERGY SPECTRUM OF THE MODEL IN THE CASE OF WEAK INTERACTION

Let us generalize the Hamiltonian proposed in Ref. 37 by taking into account the interatomic exchange interaction:

$$
\begin{aligned}
H=&-\mu\sum_{i\gamma\sigma} a^{\dagger}_{i\gamma\sigma}a_{i\gamma\sigma}+{\sum_{ij\gamma\sigma}}' t_{ij}(n)a^{\dagger}_{i\gamma\sigma}a_{j\gamma\sigma}\\
&+{\sum_{ij\gamma\sigma}}' (t'_{ij}a^{\dagger}_{i\gamma\sigma}a_{j\gamma\sigma}n_{i\bar{\gamma}}+\text{H.c.})+{\sum_{ij\gamma\sigma}}' (t''_{ij}a^{\dagger}_{i\gamma\sigma}a_{j\gamma\sigma}n_{i\gamma\bar{\sigma}}\\
&+\text{H.c.})+U\sum_{i\gamma} n_{i\gamma\uparrow}n_{i\gamma\downarrow}+U'\sum_{i\sigma} n_{i\alpha\sigma}n_{i\beta\bar{\sigma}}\\
&+(U'-J_0)\sum_{i\sigma} n_{i\alpha\sigma}n_{i\beta\sigma}+J_0\sum_{i\sigma} a^{\dagger}_{i\alpha\sigma}a^{\dagger}_{i\beta\bar{\sigma}}a_{i\alpha\bar{\sigma}}a_{i\beta\sigma}\\
&+\frac{J}{2}{\sum_{ij\gamma\gamma'\sigma\sigma'}}' a^{\dagger}_{i\gamma\sigma}a^{\dagger}_{j\gamma'\sigma'}a_{i\gamma\sigma'}a_{j\gamma'\sigma},
\end{aligned}
\tag{2.1}
$$

where μ is the chemical potential, $a^{\dagger}_{i\gamma\sigma}, a_{i\gamma\sigma}$ are the creation and destruction operators of an electron of spin σ ($\sigma=\uparrow,\downarrow$; $\bar{\sigma}$ denotes spin projection which is opposite to σ) on i site and in orbital γ ($\gamma=\alpha,\beta$ denotes two possible orbital states), $n_{i\gamma\sigma}=a^{\dagger}_{i\gamma\sigma}a_{i\gamma\sigma}$ is the number operator of electrons of spin σ and in orbital γ on i-site, $n_{i\gamma}=n_{i\gamma\uparrow}+n_{i\gamma\downarrow}$; $t_{ij}(n)$ is the effective concentration-dependent hopping integral of an electron from γ orbital of j site to γ orbital of i site (we neglect the electron hoppings between α and β orbitals), t'_{ij} (t''_{ij}) includes influence of an electron on $\bar{\gamma}$ (γ) orbital of i or j site on hopping process ($\bar{\gamma}=\beta$ if $\gamma=\alpha$, and $\bar{\gamma}=\alpha$ when $\gamma=\beta$), the primes at sums in Eq. (2.1) signify that $i=j$, U is the intra-atomic Coulomb repulsion of two electrons of the opposite spins at the same orbital (we assume that it has the same value at α and β orbitals), U' is the intra-atomic Coulomb repulsion of two electrons of the opposite spins at the different orbitals, J_0 is the intra-atomic exchange-interaction energy that stabilizes the Hund's states

forming the atomic magnetic moments, and J is the interatomic exchange interaction. The effective hopping integral $t_{ij}(n)$ is concentration dependent in consequence of taking into account the correlated hopping[40] of electron.

The peculiarities of the model described by the Hamiltonian (2.1) are taking into consideration the influence of the site occupation on the electron hoppings (correlated hopping), and the direct exchange between the neighboring sites. In this model an electron hopping from one site to another is correlated both by the occupation of the sites involved in the hopping process and the occupation of the nearest-neighbor sites. The correlated hopping, first, renormalizes the initial hopping integral (it becomes concentration and spin dependent) and, secondly, leads to a shift of the subband center that is dependent on magnetic and orbital orderings and independent on quasi-impulse. To characterize the value of correlated hopping we introduce dimensionless parameters τ [defined by $t_{ij}(n)=t_{ij}(1-n\tau)$], $\tau'=t'_{ij}/|t_{ij}|$, and $\tau_2=t''_{ij}/|t_{ij}|$, where t_{ij} is the band hopping integral.

The single-particle Green function satisfies the equation

$$
\begin{aligned}
(E+\mu)\langle\langle a_{p\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E=&\frac{\delta_{pp'}}{2\pi}+\sum_i t_{ip}(n)\langle\langle a_{i\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E+\sum_{i\sigma'} t'(ip)[\langle\langle a^{\dagger}_{p\bar{\gamma}\sigma'}a_{p\bar{\gamma}\sigma'}a_{i\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E\\
&+\langle\langle a^{\dagger}_{p\bar{\gamma}\sigma'}a_{i\bar{\gamma}\sigma'}a_{p\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E+\langle\langle a^{\dagger}_{i\bar{\gamma}\sigma'}a_{i\bar{\gamma}\sigma'}a_{i\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E+\langle\langle a^{\dagger}_{i\bar{\gamma}\sigma'}a_{p\bar{\gamma}\sigma'}a_{p\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E]\\
&+\sum_i t''(ip)[\langle\langle a^{\dagger}_{p\gamma\bar{\sigma}}a_{p\gamma\bar{\sigma}}a_{i\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E+\langle\langle a^{\dagger}_{p\gamma\bar{\sigma}}a_{i\gamma\bar{\sigma}}a_{p\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E+\langle\langle a^{\dagger}_{i\gamma\bar{\sigma}}a_{i\gamma\bar{\sigma}}a_{i\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E\\
&+\langle\langle a^{\dagger}_{i\gamma\bar{\sigma}}a_{p\gamma\bar{\sigma}}a_{p\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E]+U\langle\langle n_{p\gamma\bar{\sigma}}a_{p\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E+U'\langle\langle n_{p\bar{\gamma}\sigma}a_{p\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E+(U'-J_0)\\
&\times\langle\langle n_{p\bar{\gamma}\sigma}a_{p\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E+J_0\langle\langle a^{\dagger}_{p\bar{\gamma}\sigma}a_{p\gamma\sigma}a_{p\bar{\gamma}\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E+\sum_{i\gamma'\sigma'} J\langle\langle a^{\dagger}_{i\gamma'\sigma'}a_{p\gamma\sigma'}a_{i\gamma'\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E.
\end{aligned}
\tag{2.2}
$$

Let us consider the system at weak intra-atomic Coulomb interaction (U, U' are smaller than the bandwidth $2w=2z|t_{ij}|$ where z is the number of nearest neighbors to a site). In this case we can take into account electron-electron interactions in the Hartree-Fock approximation:

$$
\langle\langle a^{\dagger}_{i\gamma\bar{\sigma}}a_{i\gamma\bar{\sigma}}a_{j\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E\simeq\langle a^{\dagger}_{i\gamma\bar{\sigma}}a_{i\gamma\bar{\sigma}}\rangle\langle\langle a_{j\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E,
\tag{2.3}
$$

$$
\langle\langle a^{\dagger}_{i\gamma\bar{\sigma}}a_{j\gamma\bar{\sigma}}a_{i\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E\simeq\langle a^{\dagger}_{i\gamma\bar{\sigma}}a_{j\gamma\bar{\sigma}}\rangle\langle\langle a_{i\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_E.
$$

We assume that averages $\langle a^{\dagger}_{i\gamma\sigma}a_{i\gamma\sigma}\rangle=n_{\gamma\sigma}$ are independent of the number of a site, i.e. we limit ourselves to a uniform charge and electronic magnetic moment distribution. We suggest that in the case under consideration in the present paper, the orbital and antiferromagnetic orderings are destabilized by translational motion of electrons similarly to the situation observed in doped Mott-Hubbard compounds.

After the transition to Fourier representation we obtain for the Green function

$$
\langle\langle a_{p\gamma\sigma}|a^{\dagger}_{p',\gamma\sigma}\rangle\rangle_{\mathbf{k}}=\frac{1}{2\pi}\frac{1}{E-E_{\gamma\sigma}(\mathbf{k})},
\tag{2.4}
$$

where the single-particle energy spectrum is

$$
E_{\gamma\sigma}(\mathbf{k})=-\mu_{\gamma\sigma}+t_{\mathbf{k}}(n\gamma\sigma),
\tag{2.5}
$$

with the shifted chemical potential

$$
\begin{aligned}
\mu_{\gamma\sigma}=&\mu-\beta'_{\gamma}-\beta''_{\gamma\sigma}-n_{\gamma\bar{\sigma}}U-n_{\bar{\gamma}\sigma}U'-n_{\bar{\gamma}\sigma}(U'-J_0)\\
&+zJ\sum_{\sigma'} n_{\gamma\sigma'},
\end{aligned}
\tag{2.6}
$$

here the shifts of the subband centers are

$$
\beta'_{\gamma}=\frac{2}{N}\sum_{ij\sigma} t'(ij)\langle a^{\dagger}_{i\bar{\gamma}\sigma}a_{j\bar{\gamma}\sigma}\rangle,
\tag{2.7}
$$

$$
\beta''_{\gamma\sigma}=\frac{2}{N}\sum_{ij} t''(ij)\langle a^{\dagger}_{i\gamma\bar{\sigma}}a_{j\gamma\bar{\sigma}}\rangle;
\tag{2.8}
$$

and the spin- and concentration-dependent hopping integral is

$$
\begin{aligned}
t_{\mathbf{k}}(n\gamma\sigma)=t_{\mathbf{k}}\Bigg(&1-\tau n-2\tau' n_{\bar{\gamma}}-2\tau_2 n_{\gamma\bar{\sigma}}\\
&-\frac{zJ}{w}\sum_{\sigma'}\langle a^{\dagger}_{i\gamma\sigma'}a_{j\gamma\sigma'}\rangle\Bigg),
\end{aligned}
\tag{2.9}
$$

$t_{\mathbf{k}}$ is the Fourier transformant of the hopping integral t_{ij}.

The dependence of effective hopping integral on electron concentration and magnetization and a presence of the spin-dependent shift of subband center are the essential distinctions of single-particle energy spectrum of the model described by Hamiltonian (2.1) from the spectrum of the Hubbard model in the case of weak interaction.

III. FERROMAGNETISM IN THE GROUND STATE OF THE MODEL

The concentration of electrons with spin σ on γ orbital is

$$n_{\gamma\sigma}=\int_{-\infty}^{+\infty}\rho(\epsilon)f(E_{\gamma\sigma}(\epsilon))d\epsilon. \tag{3.1}$$

Here $\rho(\epsilon)$ is the density of states, $f(\epsilon)$ is the Fermi distribution function, $E_{\gamma\sigma}(\epsilon)$ is obtained from respective formula (2.5) substituting $t_{\mathbf{k}}\rightarrow\epsilon$. Let us assume the rectangular density of states,

$$\rho(\epsilon)=\frac{1}{N}\sum_{\mathbf{k}}\delta(\epsilon-\epsilon(\mathbf{k}))=\frac{1}{2w}\theta(\epsilon^2-w^2). \tag{3.2}$$

In the case of zero temperature we obtain

$$n_{\gamma\sigma}=\frac{\varepsilon_{\gamma\sigma}+w}{2w}, \tag{3.3}$$

where the value $\varepsilon_{\gamma\sigma}$ is the solution of the equation $E_{\gamma\sigma}(\varepsilon)=0$, from which we obtain $\varepsilon_{\gamma\sigma}=\mu_{\gamma\sigma}/\alpha_{\gamma\sigma}$, where $\alpha_{\gamma\sigma}=1-\tau n-2\tau' n_{\gamma}-2\tau_2 n_{\gamma\bar{\sigma}}-zJ/w\Sigma_{\sigma'}n_{\gamma\sigma'}(1-n_{\gamma\sigma'})$.

The shifts of subband centers are

$$\beta'_{\gamma}=\frac{2}{N}\sum_{ij\sigma}t'(ij)\langle a^{\dagger}_{i\bar{\gamma}\sigma}a_{j\bar{\gamma}\sigma}\rangle=-2\tau' w\sum_{\sigma}n_{\bar{\gamma}\sigma}(n_{\bar{\gamma}\sigma}-1), \tag{3.4}$$

$$\beta''_{\gamma\sigma}=\frac{2}{N}\sum_{ij}t''(ij)\langle a^{\dagger}_{i\gamma\bar{\sigma}}a_{j\gamma\bar{\sigma}}\rangle=-2\tau_2 w n_{\gamma\bar{\sigma}}(n_{\gamma\bar{\sigma}}-1). \tag{3.5}$$

From equation (3.3) we obtain for the magnetization ($m<n$)

$$\begin{aligned}m&=\sum_{\gamma}(n_{\gamma\uparrow}-n_{\gamma\downarrow})\\&=\pm 2\left(\frac{zJ}{2w}\right)^{-1/2}\left\{\frac{zJ}{8w}[8+n(4-n)]+\frac{U+J_0}{2w}+\tau_1 n\right.\\&\left.\quad+\frac{\tau_2}{2}(4-n)-1\right\}^{1/2},\end{aligned} \tag{3.6}$$

where $\tau_1=\tau+\tau'$, here we have assumed that the orbital distribution of electrons is uniform. Note that magnetization (3.6) does not depend on the parameter of intra-atomic Coulomb interaction U', which leads to the independent on magnetic moment renormalization of the chemical potential (it can be seen also from the expression for ground state energy). Similarly, in Ref. 25 it has been argued that this parameter does not play a decisive role in metallic ferromagnetism of the transition metal compounds.

The magnetization defined by Eq. (3.6) is plotted in Fig. 1 as a function of electron concentration n at different values J_0/w. These dependencies qualitatively agree with results of Ref. 23 obtained by use of the Gutzwiller variational functions method. From Fig. 1 one can see that nature of the ground state of the system strongly depends on the values of system parameters; the small changes of J_0 can lead to the transition from a paramagnetic state to a ferromagnetic one at some values of electron concentration and energy parameters (this result agrees with the results of works[24,17]); note that at some values of parameters the system can be fully polarized. The transition to ferromagnetic state is also possible with the increase of n. Similar transition with the increase of electron concentration has been found by the authors of Ref. 38.

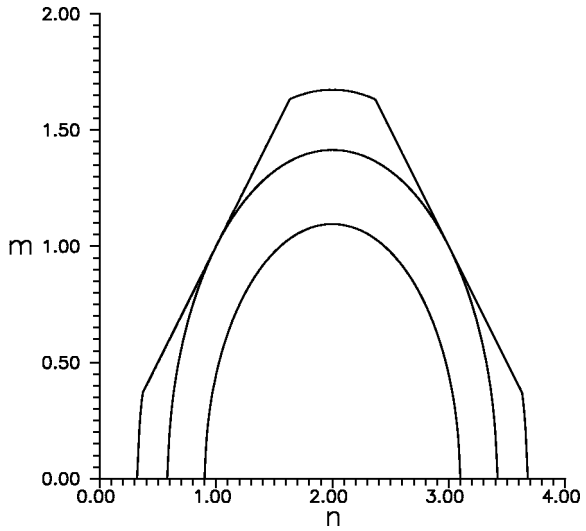

FIG. 1. The magnetization m as a function of n at $U/w=1.5$ and $\tau_1=\tau_2=0$, $zJ/w=0.1$. Upper curve corresponds to $J_0/w=0.27$, middle curve corresponds to $J_0/w=0.25$, lower one corresponds to $J_0/w=0.23$.

Taking into account correlated hopping leads to the appearance of a peculiar kinetic mechanism of ferromagnetic ordering stabilization. This mechanism is caused by the presence of the spin-dependent shift of the subband centers being the consequence of correlated hopping (which are similar to the shift of subband centers in consequence of interatomic direct exchange interaction).

The influence of correlated hopping on behavior of the system is illustrated on Fig. 2. In distinction from the two-band Hubbard model there is an asymmetry of the cases $n<2$ and $n>2$. With the increase of parameter τ_1 the region of ferromagnetic ordering moves towards larger values of electron concentration n, and with increasing τ_2—to smaller values of n. Let us also note that taking into account the correlated hopping significantly enriches the set of curves [illustrating the $m(n)$ dependencies], which qualitatively describe the experimental Slater-Pauling curves[41] for ferromagnetic alloys.

The peculiarity of degenerate-band models is taking into account Hund's exchange interaction J_0. The importance of J_0 is shown in Fig. 3. One can see that intra-atomic exchange stabilizes ferromagnetism in orbitally degenerate band (the behavior of m with the increase of J_0 qualitatively agrees with the dependence of magnetic moment on the intra-atomic correlation strength obtained in Ref. 38). To describe the real narrow-band materials we have to take into account the correlated hopping that allows to obtain the transition

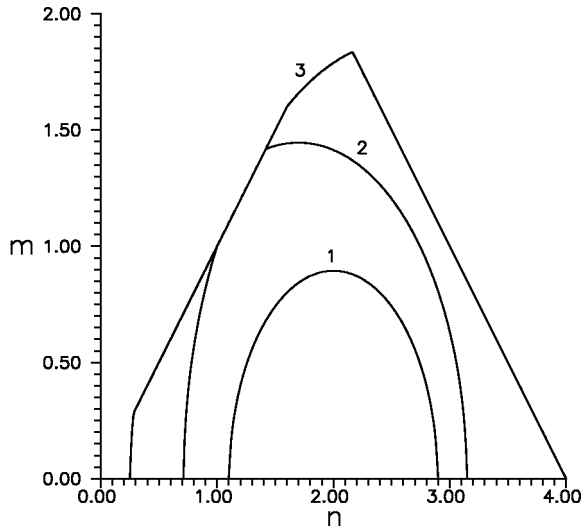

FIG. 2. The magnetization m as a function of n at $U/w=1.5$, $zJ/w=0.1$, and $J_0/w=0.22$. Curve 1 corresponds to $\tau_1=\tau_2=0$, curve 2 to $\tau_1=0$, $\tau_2=0.015$, curve 3 to $\tau_1=0.015$, $\tau_2=0$.

from paramagnetic to ferromagnetic phase at realistic values of J_0. Figure 4, which is plotted with use of Eq. (3.6) at $U/w=1.2$, $zJ/w=0.06$, and $J_0/w=0.2$, $\tau_1=0$, $\tau_2=0.15$, reproduces the behavior of the magnetization observed in the systems $Fe_{1-x}Co_xS_2$ and $Co_{1-x}Ni_xS_2$ with the change of electron concentration in $3d$ band.[42] In these crystals the same subsystem of electrons is responsible both for conductivity and for the localized magnetic-moment formation. The noted compounds have the cubic pyrite structure, then $3d$ band is split into two subbands: a doubly degenerate e_g band and a triply degenerate t_{2g} band; t_{2g} band is completely filled and e_g band is partially filled (the e_g band filling changes from 0 to 1 in the compound $Fe_{1-x}Co_xS_2$ and from 1 to 2 in the compound $Co_{1-x}Ni_xS_2$). One should describe e_g band of these compounds by a doubly orbitally degenerate model.

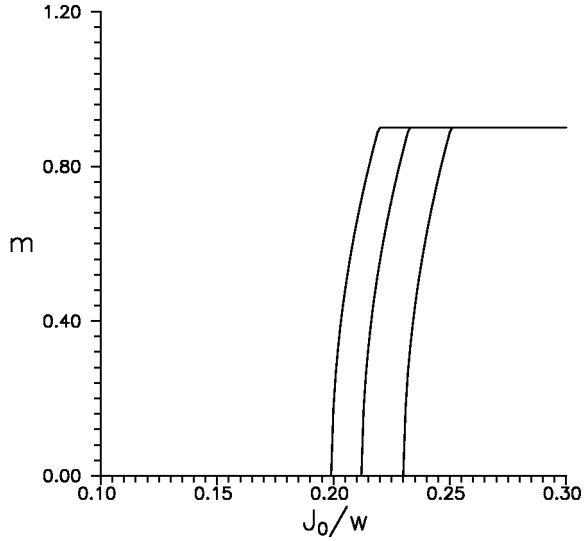

FIG. 3. The magnetization m as a function of J_0/w at $n=0.9$, $U/w=1.5$ and $zJ/w=0.1$. Left curve corresponds to $\tau_1=0$, $\tau_2=0.01$, middle curve corresponds to $\tau_1=0.01$, $\tau_2=0$, and right one corresponds to $\tau_1=\tau_2=0$.

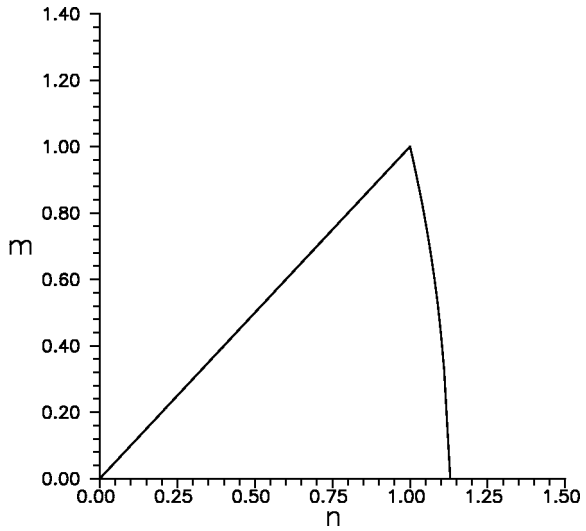

FIG. 4. The magnetization m as a function of n at $U/w=1.2$, $zJ/w=0.06$ and $J_0/w=0.2$, $\tau_1=0$, $\tau_2=0.15$.

The unusual peculiarity of the system $Fe_{1-x}Co_xS_2$ is the presence of ferromagnetic ordering at very small values of electron concentration $n=x\simeq 0.05$.[42] Ferromagnetism in this compound has been studied within a single-band model in Refs. 43 and 44. The authors of Ref. 43 have proposed an approximation for the description of $Fe_{1-x}Co_xS_2$ in the nondegenerate Hubbard model with $U=\infty$ that provides the ferromagnetic solution even at very small electron concentration (in this connection see also Ref. 44). However, in accordance with the Kanamori theory[2] at very small n we should obtain the gas limit where ferromagnetism does not occur. We also believe that the degeneracy of e_g band is essential for the description of ferromagnetic ordering in this system. Our results allow to obtain the ferromagnetism for small values of electron concentration induced by correlated hopping τ_2 in a presence of the interatomic exchange interaction (see Fig. 4). Thus, we believe that the correlated hopping mechanism in a presence of the interatomic exchange interaction allows the more natural explanation of the origin of ferromagnetism in the system $Fe_{1-x}Co_xS_2$ at very small x.

To calculate the ground-state energy of the model per site we use the formula

$$E_0=\frac{1}{2N}\sum_{\mathbf{k}\gamma\sigma}\int_{-\infty}^{+\infty}(t_{\mathbf{k}}(n)+E)J_{\mathbf{k}}^{\gamma\sigma}(E)dE. \tag{3.7}$$

Here

$$J_{\mathbf{k}}^{\gamma\sigma}(E)=\delta(E-E_{\gamma\sigma}(\mathbf{k}))\theta(-E) \tag{3.8}$$

is the spectral intensity of Green function (2.4), $\theta(-E)$ is the step-wise function. From Eq. (3.7) one can obtain for the ground state energy the expression

$$E_0=-\frac{1}{2}\sum_{\gamma\sigma}\left[\mu_{\gamma\sigma}n_{\gamma\sigma}-(1-\tau_1 n+\alpha_{\gamma\sigma})n_{\gamma\sigma}(1-n_{\gamma\sigma})w\right]. \tag{3.9}$$

Expression (3.9) can be rewritten in the form

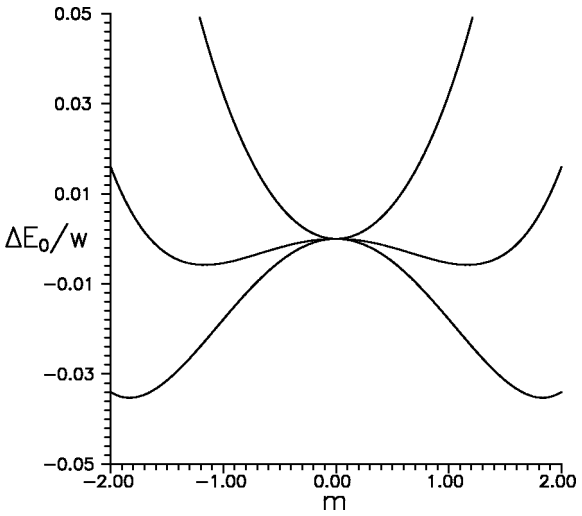

FIG. 5. The energy difference between ferromagnetic and paramagnetic ground states as a function of magnetization m at $n=1.2$, $U/w=1.2$, $zJ/w=0.2$, and $\tau_1=\tau_2=0$: upper curve corresponds to $J_0/w=0$, middle curve corresponds to $J_0/w=0.3$, and lower curve corresponds to $J_0/w=0.4$.

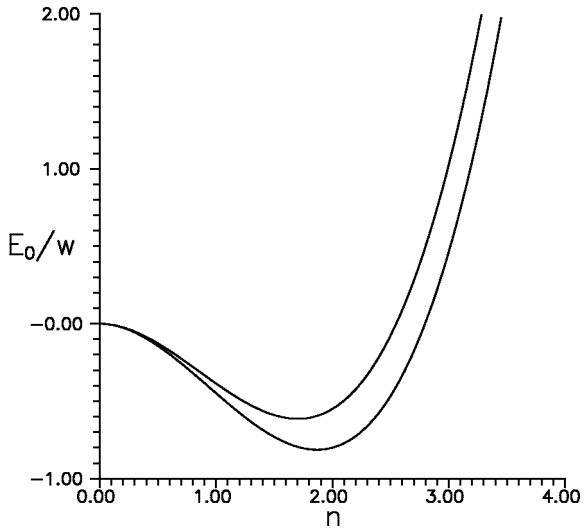

FIG. 6. The energy of ferromagnetic ground state as a function of n at $U/w=1.2$, $zJ/w=0.1$, $\tau_1=\tau_2=0$: $J_0/w=0.1$ for upper curve, and $J_0/w=0.2$ for lower one.

$$E_0=E_0^{(0)}+E_0^{(2)}+E_0^{(4)}, \tag{3.10}$$

$$E_0^{(0)}=\frac{n}{2}\left[-\mu+\frac{n}{4}\left\{U+2U'-J_0-\frac{zJ}{8}[16-(4-n)^2]\right\}-\left(1-\tau_1 n-\frac{n}{2}\tau_2\right)(4-n)\frac{w}{2}\right],$$

$$E_0^{(2)}=\left\{2[1-\tau_1 n-\tau_2(4-n)]-\frac{U}{w}-\frac{J_0}{w}+\frac{2zJ}{w}\times\left[1+\frac{n(4-n)}{8}\right]\right\}\frac{w}{8}m^2,$$

$$E_0^{(4)}=\frac{zJ}{64}m^4.$$

The position of the minimum of ground-state energy depends on values of model parameters. In Fig. 5 the energy difference ΔE_0 between paramagnetic and ferromagnetic states is plotted as a function of the magnetization. At some values of the parameters a ferromagnetic ordering with $m\neq 0$ is energetically preferred. As it has been noted above the intra-atomic exchange is an important factor leading to ferromagnetism in an orbitally degenerate band. The increase of J_0/w leads both to the increase of magnetic moment and to the decrease of the ferromagnetic ground-state energy.

The dependence of the ground-state energy of the model on the electron concentration is plotted in Fig. 6. One can see that with the increase of n Coulomb correlation becomes more and more important and the value of the ground-state energy rapidly increases, as well as at the rise of the intra-atomic Coulomb repulsion parameters. As the value of intra-atomic exchange increases the ground state energy decreases (Fig. 7); at some critical value J_0/w the transition of the system from a state of paramagnetic metal to a state of ferromagnetic metal occurs. It appears that the ferromagnetic ordering can be more favorable than the paramagnetic one in orbitally degenerate model without singularities of the density of states even if the interatomic exchange and correlated hopping are absent (in the Ref. 20 similar result is obtained only in the presence of density of states singularities). Let us also note that at increase of U the critical value of J_0, at which the transition to ferromagnetic state occurs, decreases. A qualitatively similar picture has been obtained by the authors of Ref. 15.

In Fig. 8 the energy difference between the paramagnetic and ferromagnetic states [Fig. 8(b)] and the value of magnetization [Fig. 8(a)] as functions of band filling are plotted at

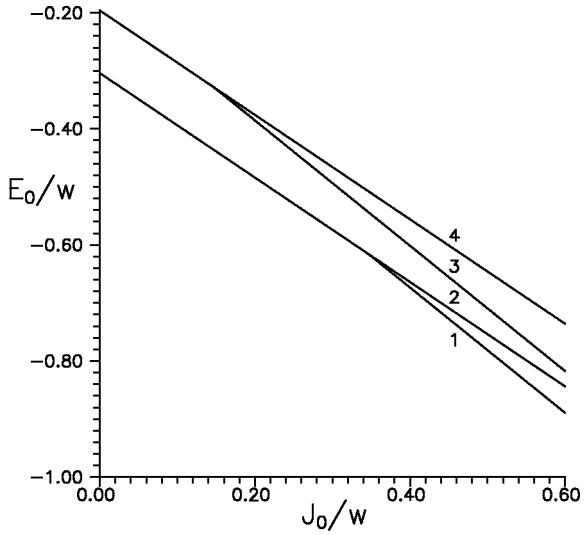

FIG. 7. The ferromagnetic (curves 1,3) and paramagnetic (curves 2,4) ground state energies as a function of J_0/w: $n=1.2$, $zJ/w=0.05$, and $\tau_1=\tau_2=0.1$; curves 1,2 correspond to $U/w=1$, curves 3,4 correspond to $U/w=1.2$.

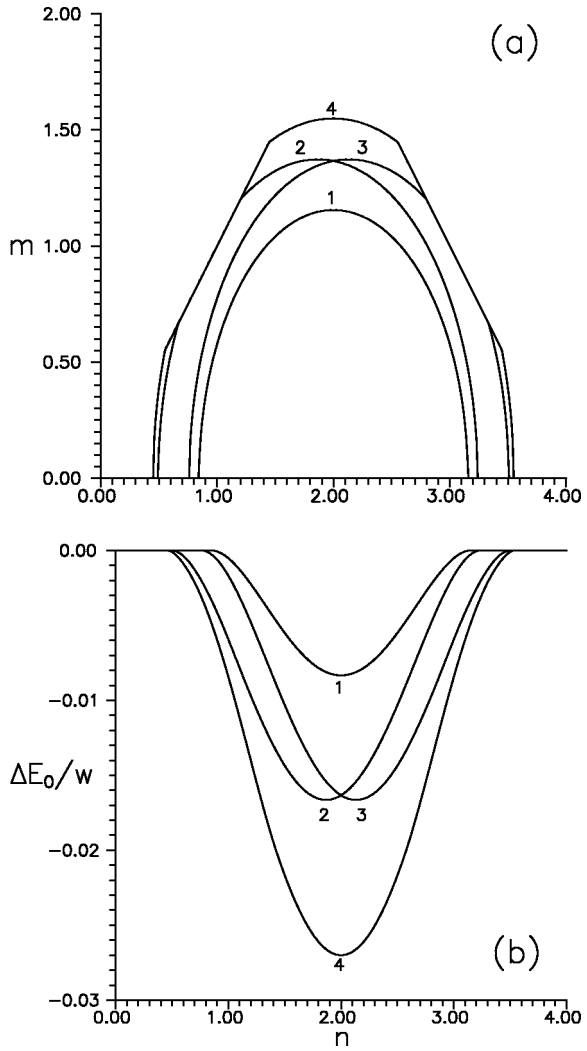

FIG. 8. The magnetization m (panel a) and the energy difference between ferromagnetism and paramagnetic ground states (panel b) as a function of n at $U/w=0.9$, $zJ/w=0.3$, and $J_0/w=0.3$: curve 1 correspond to $\tau_1=\tau_2=0$, curve 2 to $\tau_1=0$, $\tau_2=0.02$, curve 3 to $\tau_1=0.01$, $\tau_2=0$, and curves 4 to $\tau_1=0.01$, $\tau_2=0.02$.

different values of correlated hopping. Depending on the value of n (and the relation between the energy parameters) the state of the system can be paramagnetic or ferromagnetic, polarization can be full or partial. The curves 1 correspond to the case when correlated hopping is absent. With increase of n the transition from paramagnetic to ferromagnetic state occurs, in the region $n>2$ the inverse transition takes place (symmetrical behavior of the concentration dependence relative to half-filling is observed). The correlated hopping leads to a decrease of the ground-state energy, in particular, the increase of τ_1 (curves 3) has stronger influence at $n>2$ (as a result the region of ferromagnetic ordering moves towards larger values of n), the increase of τ_2 (curves 2) – at $n<2$ (the region of ferromagnetic ordering moves towards smaller values of n). These effects are the manifestation of electron-hole asymmetry being the property of the systems with correlated hoppings (see Refs. 37 and 40). At some values of correlated hopping in the system electron-hole symmetry retrieves (curves 4 in Fig. 8).

The condition of ferromagnetic ordering stability $d^2E_0/dm^2<0$ can be obtained as

$$\frac{U+J_0}{2w}+\frac{zJ}{8w}[8+n(4-n)]+n\tau_1+\frac{1}{2}\tau_2(4-n)>1. \tag{3.11}$$

From expressions for ground-state energy (3.10) and magnetization (3.6) one can see that for the values of interatomic exchange interaction $J>0$ at the point of the transition from a paramagnetic metal to a ferromagnetic metal the magnetization changes continuously, and for $J=0$ it has a jump. Namely, in the former case the transition from a paramagnetic state to a partially polarized ferromagnetic state occurs, in the later one the transition from a paramagnetic state to a fully polarized ferromagnetic state (saturated ferromagnetic state, $m=n$) is obtained. Similar results have been obtained in a single-band model.[30,45,27] Thus, taking into account the interatomic exchange interaction allows obtaining a partially polarized ferromagnetic state in two-band Hubbard model with symmetrical density of states; the partially polarized ferromagnetic state has been obtained by the authors of Ref. 23 using the special feature of the density of states.

For the case of $J=0$ from Eq. (3.11) we obtain a generalization of the Stoner criterion that takes into account the orbital degeneracy and correlated hopping

$$(U+J_0)\rho(\epsilon_F)>1-n\tau_1-\frac{1}{2}\tau_2(4-n). \tag{3.12}$$

From the condition of the minimum of ground-state energy $dE_0/dm=0$ one can also obtain the condition of partial spin polarization

$$\frac{U+J_0}{2w}+\frac{zJ}{8w}[8+n(4-n)-8m^2]+n\tau_1+\frac{1}{2}\tau_2(4-n)>1. \tag{3.13}$$

The condition of full-spin polarization ($m=n$) is

$$\frac{U+J_0}{2w}+\frac{zJ}{8w}[8+n(4-n)-8n^2]+n\tau_1+\frac{1}{2}\tau_2(4-n)>1. \tag{3.14}$$

Equations (3.11)–(3.14) coincide with the conditions that can be derived from Eq. (3.6).

From these conditions one can see that both mechanisms of correlated hopping favor ferromagnetism but their concentration dependences are different; if $\tau_2>\tau_1$ then the systems with the electron concentration $n<2$ are more favorable to ferromagnetism than the systems with $n>2$, and vice versa (see Figs. 2, 8, and 9).

In Fig. 9 the critical values of interatomic exchange are plotted as a function of band filling. The lower curve corresponds to the critical value for the partial polarization, the upper one—for the full polarization; the region below the lower curve corresponds to paramagnetic ordering of spins, between the curves—to the partial polarization, above the curves—to the full polarization of spins. It can be seen that correlated hopping essentially changes the condition of ferromagnetic ordering. Let us also note that the region of par-

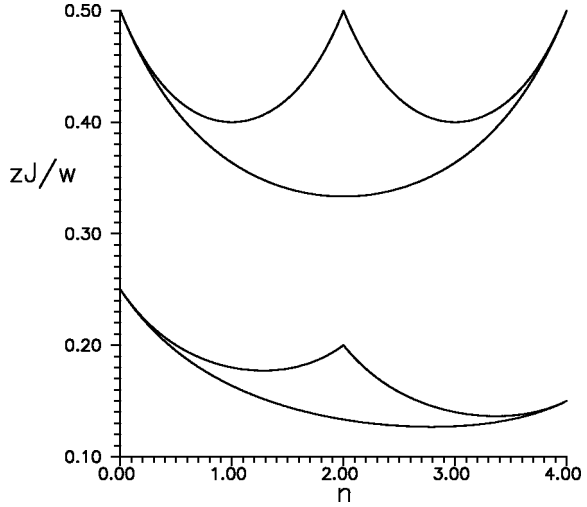

FIG. 9. The critical values of zJ/w as a function of n at $J_0/w = 0.1$. Upper curves correspond to $U/w = 0.9$, $\tau_1 = \tau_2 = 0$, lower curves to $U/w = 1.2$, $\tau_1 = 0.05$.

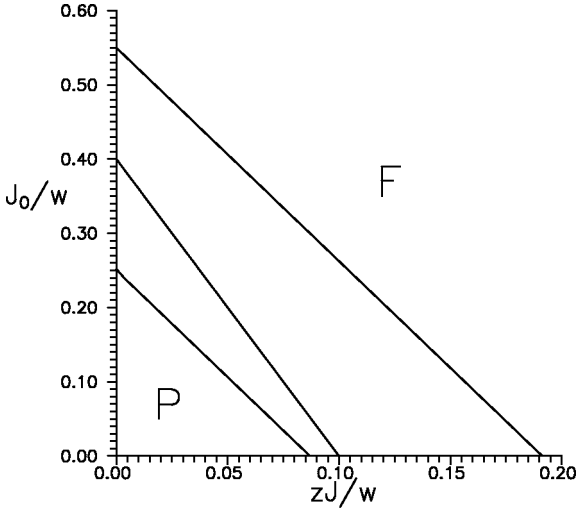

FIG. 11. The critical value of J_0/w vs zJ/w at $U/w = 1$ and $\tau_1 = \tau_2 = 0.1$ Upper curve corresponds to $n = 0.5$, middle curve to $n = 2$, and lower curve to $n = 3.5$.

tial polarization is narrowed with a deviation from half-filling (similarly to the case of nondegenerate band[27]). The maximum of critical value zJ/w corresponds to almost empty band. The cases $n = 1,3$ are the most favorable for the existence of full-spin polarization. Correlated hopping can lead to the displacement of the minimum points and to the increase or decrease of zJ/w critical values for the full polarization, i.e., to the nonequivalency of the cases $n < 2$ and $n > 2$.

In Fig. 10 the dependencies of critical values of J_0/w on electron concentration at different values of zJ/w are plotted. It is important to note that at $zJ = 0$ the critical value of J_0/w does not depend on the electron concentration. It can be explained by the following arguments: in the absence of interatomic exchange the mechanism that stabilizes ferromagnetism is a translational motion of electrons that enforces the spins of sites, involved in the hopping process, to align in parallel because of Hund's rule coupling.

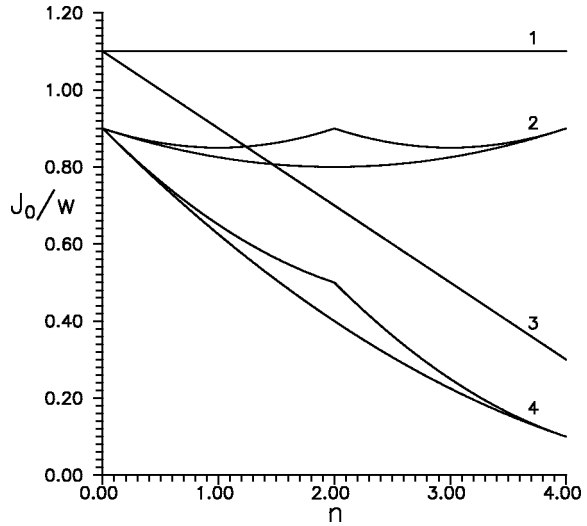

FIG. 10. The critical values of J_0/w as a function of n at $U/w = 0.9$, $\tau_2 = 0$. Curve 1 corresponds to $zJ/w = 0$, $\tau_1 = 0$, curves 2 to $zJ/w = 0.1$, $\tau_1 = 0$, curve 3 to $zJ/w = 0$, $\tau_1 = 0.01$, and curves 4 correspond to $zJ/w = 0.1$, $\tau_1 = 0.01$.

Note also the essential difference of the situation where the system is described by nonzero values of correlated hopping: since correlated hopping renormalizes the bandwidth and makes it dependent on the concentration, the behavior of the critical value of J_0/w becomes asymmetrical relative to half-filling.

The critical value of J_0/w as a function of zJ/w is plotted on Fig. 11. As one can see the increase of zJ/w significantly decreases the critical value of J_0/w (in the same way the correlated hopping does). It shows the importance of taking into account the interatomic exchange and correlated hopping for the description of ferromagnetism in the systems with orbital degeneracy. The inverse dependence of critical values of zJ/w and J_0/w (which indicates the destabilization of ferromagnetic ordering at the increase of J_0/w) has been obtained in Ref. 24 with use of the exact diagonalization method for the even number of sites in one-dimensional chains, but that result depends sensitively on the number of lattice sites and the boundary conditions.

IV. CONCLUSIONS

In this paper we have investigated the ground state of a doubly orbitally degenerate model. Taking into consideration the orbital degeneracy allows to analyze the influence of intra-atomic exchange interaction (Hund's rule coupling), which is responsible for the formation of local magnetic moments, on the possibility of ferromagnetism realization. Besides the diagonal matrix elements of electron-electron interactions the model includes the off-diagonal ones—correlated hopping integrals, which describe the influence of site occupancy on the hopping of electrons. The model under consideration also includes the interatomic exchange interaction J.

The study of the model ground state, carried out in this work, shows that the stability of ferromagnetism strongly

depends on the model parameters. In particular, it has been found that the relationship between correlated hopping parameters determines the criterion of ferromagnetism. At the values of correlated hopping parameters $\tau_2 > \tau_1$ in the system with concentration of electrons $n < 2$ the situation for ferromagnetic ordering is more favorable than for the system with $n > 2$; at $\tau_2 < \tau_1$ the opposite behavior is obtained. At some values of correlated hopping parameters the retrieval of the electron-hole symmetry is possible. Taking into account the correlated hopping leads to the appearance of specific mechanism that stabilizes ferromagnetic ordering, and this is due to the spin-dependent shift of the subband centers. In the absence of interatomic exchange interaction the ferromagnetic ordering is stabilized by the translational motion of electrons between sites with ''atomic ferromagnetism'' formed by Hund's rule coupling.

It is important to note that the transition of the system from paramagnetic to ferromagnetic state can occur at the values of interaction parameters, which are of the same order as that of bandwidth, and with density of states without peculiarities. The important role for the ferromagnetism stabilization in weak-interaction regime ($U < 2w$) is played by the intra-atomic and interatomic exchange interactions as well as correlated hopping that allows to describe the metallic paramagnetic-ferromagnetic transition with realistic relationship between above-mentioned exchange interactions.

For the values of interatomic exchange interaction $J > 0$ at the point of the transition from a paramagnetic metal to a ferromagnetic metal the magnetization changes continuously, and for $J = 0$ it has a jump, namely, in the former case the transition from a paramagnetic state to a partially polarized ferromagnetic state occurs, in the later one the transition from a paramagnetic state to a fully polarized ferromagnetic state (saturated ferromagnetic state) is obtained.

The obtained dependencies of magnetization on concentration of electrons qualitatively describe the experimental Slater-Pauling's curves for ferromagnetic alloys. At some values of the model parameters the experimental dependence of magnetization for the systems $Fe_{1-x}Co_xS_2$ and $Co_{1-x}Ni_xS_2$ with changing electron concentration in e_g band is reproduced theoretically. The correlated hopping mechanism of ferromagnetism stabilization allows to explain the ferromagnetism in the systems $Fe_{1-x}Co_xS_2$ at small concentrations $x \simeq 0.05$.

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