Ground state energy of metallic ferromagnet in a generalized Hubbard model

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Abstract

In the present work ferromagnetic ordering in the Hubbard model generalized by taking into account the inter-atomic exchange interaction and correlated hopping in partially filled narrow band is considered. In the case of weak electron-electron interaction the ground state energy and condition of ferromagnetic state realization are found by using the Green function technique. The obtained results indicate the important role of correlated hopping.

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1 Introduction

The problem of the origin of the metallic ferromagnetism, in spite of the variety of theoretical attempts to solve it, still remains open. The simplest model at the basis of which one has tried to explain the physics of ferromagnetism in 3d transition metals is the Hubbard model that describes itinerant electrons in a single non-degenerate band interacting via on-site Coulomb repulsion $U$. The interplay between the kinetic energy of electrons and Coulomb interaction strongly influences the magnetic ordering. In some approximations the ferromagnetic solution in this model was obtained for large values of intra-atomic Coulomb interaction $U$; in this connection note the Nagaoka’s theorem (the ferromagnetism in half-filled band with single electron or single hole at $U \to \infty$). The problem of metallic ferromagnetism is studied intensively during last years (for recent reviews see Ref. [5, 6]) however the undivided opinion is not reached so far.

Nowadays we can distinguish a few ways to obtain the ferromagnetic solution: first is to stay in the framework of the Hubbard model but to use a special lattice geometry and density of states; second is to go beyond the Hubbard model including the band degeneracy (note that real ferromagnetic materials have the orbital degeneracy); and third is to take into account other matrix elements of electron-electron interactions in addition to the intra-atomic Coulomb interaction. We believe that third way can give the qualitative explanation of the magnetic properties of narrow band materials although the construction of consistent theory of metallic ferromagnetism should include the orbital degeneracy and real density of states. The aim of this article is to study the influence of so-called ”off-diagonal” matrix elements of electron-electron interaction on the condition
of ferromagnetism stabilization. In spite of the fact that the magnitudes of the neglected in the Hubbard Hamiltonian terms are small in comparison with on-site repulsion or band hopping integral but they can essentially influence (assist or suppress) the ferromagnetic ordering. The importance of some of these matrix elements was pointed out in many works \cite{7}-\cite{9}; here we note the special role of the direct exchange interaction and correlated hopping (taking into account of the inter-atomic density-density Coulomb interaction which play essential role in charge ordering goes beyond the goal of this article).

Last ten years the problem of importance of exchange interaction again is topical especially in the works \cite{10}-\cite{13}, where it was concluded that the nearest-neighbour exchange interaction \( J \) plays a fundamental role for the existence of ferromagnetism. In the cited papers the condition for ferromagnetism in the framework of mean-field theory has been obtained; also the exact diagonalization solutions for one-dimensional system with different band-fillings have been constructed and compared with results of mean-field theory \cite{13}.

The exact criteria for the arising of ferromagnetism in the partial case of the model with correlated hopping have been derived by Strack and Vollhardt \cite{14}. These criteria also show the significance of the direct exchange interaction. Vollhardt and co-workers \cite{15, 16} within dynamical mean-field theory with the asymmetrical density of states having a peak near the band edge pointed out that already small direct exchange can stabilize the ferromagnetic ordering.

As mentioned there is an additional mechanism which can favour ferromagnetic ordering. The "off-diagonal" matrix elements of Coulomb interaction of electrons include the term that describes the density-dependent hopping – so-called correlated hopping. The importance of correlated hopping and its considerable place in the descriptions of metal-insulator transition and superconductivity are known \cite{18}-\cite{25}. So we expect that taking into account the correlated hopping can give more clear understanding of the metallic ferromagnetism. The attempt to take into account the influence of correlated hopping has been done by Hirsch in Ref. \cite{13}. It has been shown that correlated hopping can give rise to the asymmetry of the condition of ferromagnetic ordering (to enhance the tendency to ferromagnetism of electron-like versus hole-like carriers for small \( U \), and have the opposite effect for large \( U \)). In our previous work we also have pointed out the importance of electron-hole asymmetry in the theory of metal-insulator transition and metallic ferromagnetism \cite{18}. It is important that correlated hopping give rise to concentration dependency of hopping integral.

The purpose of this paper is to investigate the possibility of ferromagnetic ordering and to study both the influence of correlated hopping of electrons and direct exchange interaction on the condition of ferromagnetism. The paper is organized as follows. In the Section 2 the Hamiltonian of the model with electron-hole asymmetry is formulated. In the Section 3 the single-particle Green function and energy spectrum are found in the case of weak electron-electron interaction. In the Section 4 the ground state energy of the model and the criteria of ferromagnetism stabilization are calculated and discussed. Section 5 is devoted to the conclusions.
2 The model Hamiltonian

In accordance with the works \[7, 17, 18\] we take into account in the Hamiltonian the matrix elements $J(ijji) = J$, $J(ikjk) (k \neq i, k \neq j)$, $J(iiij)$, where

$$J(ijkl) = \int \int \phi^*(r - R_i) \phi(r - R_k) \frac{e^2}{|r - r'|} \phi^*(r' - R_j) \phi(r' - R_l) dr dr'. \tag{1}$$

The model Hamiltonian takes the following form \[18\]

$$H = - \mu \sum_{i\sigma} a^+_i \sigma a_i \sigma + \sum_{ij\sigma} (t_{ij} + nT_1(ij))a^+_i \sigma a_j \sigma + \sum_{ij\sigma} (T_2(ij)a^+_i \sigma a_j \sigma + h.c.)$$

$$+ U \sum_i n_{i\uparrow} n_{i\downarrow} + \frac{J}{2} \sum_{ij\sigma'\sigma} a^+_i \sigma a^+_j \sigma' a_i \sigma a_j, \tag{2}$$

where $a^+_i \sigma$, $a_i \sigma$ are creation and destruction operators of electron on site $i$, $\sigma = \uparrow, \downarrow$, $n_{i\sigma} = a^+_i \sigma a_i \sigma$, $n = \langle n_{i\uparrow} + n_{i\downarrow} \rangle$, $\mu$ is the chemical potential, $t_{ij}$ is the band hopping integral of an electron from site $j$ to site $i$, $U$ is the intra-atomic Coulomb repulsion and $J$ is the exchange integral for the nearest neighbours,

$$T_1(ij) = \sum_{k \neq i \neq j} J(ikjk)$$

and $T_2(ij) = J(iiij)$ are the parameters of correlated hopping of electrons. The prime on the sums in Eq. (2) signifies that $i \neq j$.

The peculiarities of the model described by the Hamiltonian (2) is taking into consideration the influence of the site occupation on the electron hopping (correlated hopping), and the exchange integral. In this model an electron hopping from one site to another is correlated (in contrast to similar generalized Hubbard models) both by the occupation of the sites involved in the hopping process (with the hopping integral $T_2$) and the occupation of the nearest-neighbour sites (with the hopping integral $T_1$) which we took into account by means of the Hartree-Fock approximation.

Thereby the correlated hopping, firstly, renormalizes the initial hopping integral (it becomes concentration- and spin-dependent) and, secondly, leads to an independent on quasiimpulse shift of the band center, dependent on magnetic ordering. Taking into account the quantity of second order $J$ is in principle necessary to describe ferromagnetism in this model.

To characterize the value of the correlated hopping we introduce dimensionless parameters $\tau_1 = \frac{T_1(ij)}{|t_{ij}|}$, $\tau_2 = \frac{T_2(ij)}{|t_{ij}|}$.

3 The Green function and energy spectrum in case of weak interaction

The single-particle Green function satisfies the equation

$$(E - \mu) \langle\langle a_{p\sigma} | a_{p'\sigma}^+ \rangle\rangle_E = \frac{\delta_{pp'}}{2\pi} + \sum_i t_{ip}(n) \langle\langle a_{i\sigma} | a_{p'\sigma}^+ \rangle\rangle_E$$
Let us consider the system at weak intra-atomic Coulomb interaction ($U < 2w$, $w = z|t_{ij}|$ is the half of bare bandwidth, $z$ is the number of nearest neighbours to a site). Then we can take into account the electron-electron interactions in the Hartree-Fock approximation:

$$\langle\langle a_{i\sigma}^+ a_{i\sigma} | a_{p'\sigma}^+ \rangle\rangle_E \approx \langle a_{i\sigma}^+ a_{i\sigma} \rangle \langle\langle a_{j\sigma} | a_{p'\sigma}^+ \rangle\rangle_E; \quad (4)$$

$$\langle\langle a_{i\sigma}^+ a_{j\sigma} a_{i\sigma} | a_{p'\sigma}^+ \rangle\rangle_E \approx \langle a_{i\sigma}^+ a_{j\sigma} \rangle \langle\langle a_{i\sigma} | a_{p'\sigma}^+ \rangle\rangle_E.$$

We assume that averages $\langle a_{i\sigma}^+ a_{i\sigma} \rangle = n_{\sigma}$ are independent of the number of the site (the uniform distribution of electronic density and magnetic moments is assumed).

The approximation (4) corresponds to the condition of weak electron-electron interaction, where doubly occupied states can be taken into account by the effective mean field depending on the electron concentration and correlated hopping integral.

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

$$\langle\langle a_{p\sigma} | a_{p'\sigma}^+ \rangle\rangle_k = \frac{1}{2\pi} \frac{1}{E - E_\sigma(k)}, \quad (5)$$

where the energy spectrum is

$$E_\sigma(k) = -\mu + \beta_\sigma + n_\delta U - zn_\sigma J + t(n\sigma)\gamma(k); \quad (6)$$

here the spin-dependent shift of the band center is

$$\beta_\sigma = \frac{2}{N} \sum_{ij} T_2(ij) \langle a_{i\sigma}^+ a_{j\bar{\sigma}} \rangle, \quad (7)$$

$$\gamma(k) = \sum_{\mathbf{R}} e^{i\mathbf{k}\mathbf{R}} \quad \text{(the sum goes over the nearest neighbours to a site) and the spin and concentration dependent hopping integral is}$$

$$t(n\sigma) = t + nT_1 + 2n_\delta T_2 + J \sum_{\sigma'} \langle a_{i\sigma}^+ a_{j\sigma'} \rangle. \quad (8)$$

From the energy spectrum (6) by neglecting the last term in (8) we obtain the result of papers [17, 18]; neglecting the correlated hopping $T_1(ij)$ - the result of Ref. [13].

The dependences of effective hopping integral on electron concentration and magnetization, a presence of the spin-dependent shift of band center are the essential distinctions of single-particle energy spectrum of the model described by Hamiltonian (3) from the spectrum of the Hubbard model in the case of weak interaction.

The concentration of electrons with spin $\sigma$ is

$$n_\sigma = \int_{-\infty}^{+\infty} \rho(\epsilon)f(\epsilon)d\epsilon. \quad (9)$$
Here $\rho(\epsilon)$ is the density of states, $f(\epsilon)$ is the Fermi distribution function. Let us assume the rectangular density of states:

$$
\rho(\epsilon) = \frac{1}{N} \sum_k \delta(\epsilon - \epsilon(k)) = \frac{1}{2w} \theta(\epsilon^2 - w^2). \tag{10}
$$

In the case of zero temperature we obtain:

$$
n_\sigma = \frac{\varepsilon_\sigma + w}{2w}, \tag{11}
$$

here the value $\varepsilon_\sigma$ is the solution of the equation $E_\sigma(\varepsilon) = 0$ from which we obtain $\varepsilon_\sigma = \mu_\sigma - \beta_\sigma + zn_\sigma J - n_\sigma U$ and $\alpha_\sigma = 1 - \tau_1 n - 2\tau_2 n_\sigma - \frac{zJ}{w} \sum n_{\sigma'}(1 - n_{\sigma'})$.

The system parameters are related by the equation

$$
zJ + U + \frac{\beta_- - \beta_+}{m} = 2w \left( 1 - \tau_1 n - \tau_2 - \frac{zJ[n(2 - n) - m^2]}{2w} \right). \tag{12}
$$

The shift of band center is obtained from

$$
\beta_\sigma = \frac{2}{N} \sum_{ij} T_2(ij) \langle a_{i\sigma}^+ a_{j\sigma} \rangle = -\tau_2 w n_\sigma (n_\sigma - 1). \tag{13}
$$

One can see that

$$
\beta_- - \beta_+ = 2\tau_2 m w (1 - n). \tag{14}
$$

In particular, the concentration dependent shift (14) of the centers of spin-up and spin-down electron bands at $n < 1$ is positive, at $n > 1$ is negative. In this manner we obtain the condition for the equilibrium value of magnetization:

$$
1 - \tau_1 n - \tau_2 (2 - n) - \frac{U}{2w} = \frac{zJ}{2w} \left[ 1 + n(2 - n) - m^2 \right]. \tag{15}
$$

This result will be compared with result of next section.

### 4 Ground state energy

To calculate the ground state energy of the model we use the formula

$$
E_0 = \frac{1}{2N} \sum_{k, \sigma}^{+\infty} \int_{-\infty}^{+\infty} (t_k(n) + E) J^\sigma(E) dE. \tag{16}
$$

Here

$$
J^\sigma(E) = \frac{\delta(E - E^\sigma(k))}{1 + \exp \frac{E - \mu_\sigma}{kT}} \tag{17}
$$

is the spectral intensity of Green function (11). From Eq. (16) one can obtain for the ground state energy the expression

$$
E_0 = \frac{1}{2} \sum_{\sigma} (-\mu_\sigma n_\sigma - (1 - \tau_1 n + \alpha^\sigma) n_\sigma (1 - n_\sigma) w), \tag{18}
$$
which can be rewritten in the form
\[ E_0 = E_0^{(0)} + E_0^{(2)} + E_0^{(4)}, \]  
\[ E_0^{(0)} = \frac{n}{2} \left( -\mu + \frac{U}{2} n + \frac{zJ n(2-n)}{4} - (1 - (\tau_1 + \tau_2)(2-n)) \right), \]  
\[ E_0^{(2)} = \left( 1 - n \tau_1 - \tau_2 (2-n) - \frac{U}{2w} - \frac{zJ}{4w} (1 + n(2-n)) \right) \frac{w}{2} m^2, \]  
\[ E_0^{(4)} = \frac{zJ}{8} m^4. \]

One can see that the condition for equilibrium value of magnetization obtained from expression \( dE_0/dm = 0 \) is equivalent to Eq. (15). The position of the minimum of ground state energy depends on values of model parameters. In Fig.1 the energy difference between paramagnetic and ferromagnetic states is plotted as a function of the magnetization. From this plot we can see that at some values of the parameters a ferromagnetic ordering occurs with increase of inter-atomic exchange integral. The values of magnetization at which the ground state energy of the system has a minimum depends on magnitude of inter-atomic exchange. The condition of ferromagnetic ordering stability \( d^2E_0/dm^2 < 0 \) can be obtained as
\[ \frac{zJ}{2w} > \frac{1 - n \tau_1 - \tau_2 (2-n) - \frac{U}{2w}}{1 + n(2-n)}. \]  
(20)

For the case of \( zJ/2w = 0 \) from Eq. (20) we obtain a generalization of the Stoner criterion which takes into account the correlated hopping
\[ U \rho(\epsilon_F) > 1 - n \tau_1 - \tau_2 (2-n). \]  
(21)

From the condition of the minimum of ground state energy \( dE_0/dm = 0 \) one can also obtain for the value of exchange integral at fixed magnetization \( m \)
\[ \frac{zJ}{2w} = \frac{1 - n \tau_1 - \tau_2 (2-n) - \frac{U}{2w}}{1 + n(2-n) - m^2}. \]  
(22)

The condition of full spin polarization \( (m = n) \) is
\[ \frac{zJ}{2w} > \frac{1 - n \tau_1 - \tau_2 (2-n) - \frac{U}{2w}}{1 + 2n(1-n)}. \]  
(23)

From Eq. (20)-(23) one can conclude: the competition between correlated hopping parameters \( \tau_1 \) and \( \tau_2 \) determines more favourable situation for the ferromagnetic ordering. For example, if \( \tau_2 > \tau_1 \) then the systems with the electron concentration \( n < 1 \) are more favourable to ferromagnetism than the systems with \( n > 1 \) and vice versa (see Fig. 2, 3; the values above the line correspond to the ferromagnetic state of the system, below the line - to the paramagnetic one), but the correlated hopping parameter \( \tau_2 \) influences the condition of ferromagnetic ordering stronger than the parameter \( \tau_1 \) (see Fig. 4). Note also that the region of partial spin polarization is narrowed with a deviation of electron concentration from half-filling (see Fig. 2, 5, 6).

One can see that in the less than half-filled band, correlated hopping leads to the stabilization of ferromagnetism as well as inter-atomic exchange interaction and intra-atomic Coulomb interaction; the larger is the electron concentration \( n \), the smaller is the
critical value of the exchange integral for the occurrence of ferromagnetism. These our results are in accordance with the results of Ref. [13].

The magnetization found from the condition of minimum of the ground state energy is written as

$$m = \left( \frac{zJ}{2w} \right)^{\frac{1}{2}} \left( \frac{zJ}{2w} \frac{1+n(2-n)}{1+n(2-n)} + \frac{U}{2w} + \tau_1 n + \tau_2 (2-n) - 1 \right)^{\frac{1}{2}},$$ (24)

The magnetization defined by Eq. (24) is plotted as a function of $zJ/w$ for different values of $U/w$ in the Fig. 7. In the ferromagnetic state the magnetization rapidly increases with $zJ/w$ and reach its maximum value at relatively small values of exchange integral what indicates the importance of inter-atomic exchange interaction for ferromagnetism of the system. The dependence of the magnetization on concentration of electron is shown in Fig. 8. At fixed values of model parameters the change of electron concentration (doping of the system) can induce ferromagnetism.

In Fig. 9 the dependence of paramagnetic and ferromagnetic ground state energies of the system on $zJ/w$ is plotted for different values of $n$. For all values of the electron concentration at sufficiently large values of $zJ/w$ the energy of the ferromagnetic state lies much lower than the paramagnetic one. The position of the transition point depends on values of $U$ and $\tau_1, \tau_2$. With the increase of these parameters the critical value of inter-atomic exchange interaction becomes smaller (the influence of the Coulomb repulsion is illustrated in Fig. 10).

The energy difference between the paramagnetic and ferromagnetic states, the value of magnetization as functions of band filling are plotted in Fig. 11. Depending on the value of $n$ the state of the system can be para- or ferromagnetic. For the zero values of correlated hopping parameters (the curve 1 on Fig. 11(a)) in the system with increase of $n$ the ferromagnetic order occurs. Note that with further increase of $n$ the system becomes fully spin-polarized (see fig. 11(b)) however in almost half-filled band the spin polarization is partial again. In the more then half-filled band (in consequence of the electron-hole symmetry of this case) the noted magnetic behaviour repeats in inverse sequence. In the case of non-zero values of correlated hopping parameters (the curves 2,3,4 on Fig. 11(a)) we have the valuable decrease of ferromagnetic ground state energy comparing to paramagnetic one; the influence of correlated hopping $\tau_1$ is stronger at $n > 1$ and the influence of $\tau_2$ is stronger at $n < 1$. In the case of equal values of $\tau_1$ and $\tau_2$ the electron-hole symmetry retrieves (the curve 4 on Fig. 11(a)).

5 Conclusions

Although the consistent theory of ferromagnetic ordering in transition metal compounds can be constructed only in the model including the orbital degeneracy of the band, the qualitative character of observed properties can be interpreted in the framework of generalized model of non-degenerate band. We have studied the ferromagnetism in this model of at weak intra-atomic interaction. The peculiarity of the studied model is taking into account besides the intra-atomic Coulomb and inter-atomic exchange interactions the additional terms describing the interaction of the hopping electrons with other electrons (correlated hopping). In the considered case of weak electron-electron interaction the inter-atomic exchange interaction leads to the transition from a paramagnetic to a ferromagnetic state of the system.
The critical value of exchange interaction strongly depends on model parameters. The correlated hopping causes the spin-dependent shift of the band center which can lead to the ferromagnetism. This our result is in accordance with the results of Refs. [11, 13]. In distinction from the model considered in Ref. [13] our Hamiltonian contains additional mechanism of correlated hopping which is shown to favour the ferromagnetic ordering. The intra-atomic Coulomb interaction is the factor favouring the ferromagnetic ordering but this interaction itself can not stabilize the ferromagnetic ordering if \( U < 2w \) (see Fig.1). The ground state energy has strong filling dependence and its behaviour can be changed by rise of the correlated hopping parameters.

To conclude, in the case of weak electron-electron interaction the inter-atomic exchange interaction plays a substantial role in stabilization of the ferromagnetic ordering; the intra-atomic Coulomb interaction and the correlated hopping also favour ferromagnetism. The non-equivalence of less then half-filled and more than half-filled cases (concerning the condition of ferromagnetism realization) is shown which is characteristic for the models with correlated hopping of electrons.

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Figure captions

Fig.1 The energy difference between paramagnetic and ferromagnetic states $E(m)_w$ as a function of the magnetization $m$ at $U_w = 1$, $\tau_1 = \tau_2 = 0.2$ and $n = 0.7$. Upper, middle and lower curves correspond to $\frac{zJ}{w} = 0$, $\frac{zJ}{w} = 0.3$ and $\frac{zJ}{w} = 0.5$ respectively.

Fig.2 Ground state phase boundaries at $U_w = 0.8$, $\tau_1 = 0.15$ and $\tau_2 = 0.2$: curve 1 corresponds to the paramagnetic case, curves 2,3 - the cases of partially spin polarization, 50 and 75 percents respectively, curve 4 corresponds to the case of full spin polarization.

Fig.3 The critical value of the exchange integral as a function of $\tau_1 = 0.15$, $\tau_2 = 0.2$: curve 1 corresponds to $n = 0.5$, curve 2 corresponds to $n = 1$ and curve 3 corresponds to $n = 1.5$.

Fig.4 The critical value of the exchange integral as a function of electron concentration $n$ for given magnetization at $\tau_1 = 0.15$, $\tau_2 = 0.2$ (solid lines) and $\tau_1 = 0.2$, $\tau_2 = 0.15$ (dashed lines), $U_w = 0.8$: curves 1,2,3 and 4 correspond to $m = 0.1$, $m = 0.5$, $m = 0.7$ and $m = 0.9$ respectively.

Fig.5 Lines of constant magnetization for half-filled band: curves 1,2 and 3 correspond to the cases $m = 0$, $m = 0.7$ and $m = 1$ respectively.

Fig.6 Lines of constant magnetization for quarter-filled band: curves 1,2 and 3 correspond to the cases $m = 0$, $m = 0.7$ and $m = 1$ respectively.

Fig.7 The magnetization $m$ as a function of $\frac{zJ}{w}$ at $\tau_1 = \tau_2 = 0.1$ and $n = 0.8$. Curves 1, 2 correspond to $\frac{U}{w} = 1.2$ and $\frac{U}{w} = 0.7$ respectively.

Fig.8 The magnetization $m$ as a function of $n$ at $\frac{U}{w} = 1.2$ and $\tau_1 = \tau_2 = 0.1$. Solid curve corresponds to $\frac{zJ}{w} = 0.3$, dashed curve corresponds to $\frac{zJ}{w} = 0.6$.

Fig.9 The paramagnetic (dashed line) and ferromagnetic (solid line) ground state energies of the system as a function of $\frac{zJ}{w}$ at $\frac{U}{w} = 0.5$, $\tau_1 = \tau_2 = 0.3$. Upper curves correspond to $n = 1.2$, lower ones correspond to $n = 0.7$.

Fig.10 The ground state energy as a function of $\frac{zJ}{w}$ at $\tau_1 = \tau_2 = 0.2$ and $n = 0.8$. Curves 1,2 and 3 correspond to $\frac{U}{w} = 1$, $\frac{U}{w} = 0.6$ and $\frac{U}{w} = 0.2$ respectively.

Fig.11 The energy difference between the paramagnetic and ferromagnetic states $E(m)_w$ (a) and the value of magnetization $m$ (b) as functions of band filling $n$ at $\frac{U}{w} = 1.2$, $\frac{zJ}{w} = 0.45$. Curves 1, 2, 3 and 4 correspond to the cases $\tau_1 = \tau_2 = 0$, $\tau_1 = 0$, $\tau_2 = 0.05$, $\tau_1 = 0.05$, $\tau_2 = 0$ and $\tau_1 = \tau_2 = 0.05$ respectively.
