Chemical potential of the generalized Hubbard model with correlated hopping

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In the present paper we study chemical potential of the generalized Hubbard model with correlated hopping. The peculiarity of the model in comparison with similar generalized Hubbard models is the concentration dependence of hopping integrals. Chemical potential as a function of the model energy parameters, electron concentration and temperature is found. It is shown that correlated hopping and temperature changes essentially the chemical potential location; these dependencies differ strongly at different values of the electron concentration.

I. INTRODUCTION

One of the simplest models, which is used for a description of strongly correlated electron systems (in particular, transition metal compounds, polymers, fullerenes C_{60}, high-temperature superconductors, heavy fermion substances) is the Hubbard model [1] (for reviews see papers [2]-[4]). Hubbard first showed that taking into account the intra-atomic Coulomb repulsion $U$ of two electrons essentially modifies the band energy spectrum and plays the main role in the formation of electrical and magnetic properties of the material with narrow energy bands. The ideas of “lower” and “upper” Hubbard bands, introduced in this paper, proved to be useful both for our understanding of physics of correlation effects in narrow energy bands and for the interpretation of experimental data for narrow band materials, in particular high-$T_c$ superconductors. It should be noted that progress in experimental techniques, such as photoemission and x-ray absorption spectroscopy, has recently confirmed and enabled direct measurements of these theoretically predicted Hubbard bands [5]-[7].

However, theoretical analyses, on the one hand, and available experimental data, which argue an electron-hole asymmetry, on the other hand, point out the necessity of the Hubbard model generalization by taking into account correlated hopping (electron-hole asymmetry is a peculiar property of the Hubbard model). Really, in some compounds (e.g. see the estimation in Refs. [8]-[14]) the matrix elements of electron-electron interactions describing inter-site hoppings of electrons (correlated hopping) are of the same order that the hopping integral or on-site Coulomb repulsion; also, the results of the first principles calculations performed by Hirsch [15] show that hopping integrals of electrons between neighbouring ions can depend strongly on the instantaneous charge occupation of the two atoms involved in the hopping process (in contrast to the Hubbard model where inter-site hoppings of electrons do not correlate by the occupation of these sites). On the other hand, the experimental observations show electron-hole asymmetry of properties of strongly correlated electron systems. In particular, transition metal oxides exhibit the electron-hole asymmetry of conductivity [16], transition 3d-metals have the electron-hole asymmetry of cohesive energy [17,18]. In this connexion, the experimentally observed [19,20] electron-hole asymmetry of superconducting properties of high-temperature superconductors should be also noted.

Consequently, in recent years the generalized Hubbard model with correlated hopping has been used widely to describe strongly correlated electron systems [21]-[36]; the electron-hole asymmetry is a property of such a generalized Hubbard model as a result of the dependence of the hopping integral on the occupation of the sites involved in the hopping process.

Generalized Hubbard model has been used for an investigation of metallic ferromagnetism in narrow energy bands [22]-[23]. In particular, a generalization of Nagaoka’s theorem has been proved in paper [22], and it has been shown [23] that in strong coupling regime the model strongly favours ferromagnetism for electron concentration $n > 1$ versus electron concentration $n < 1$, as observed in transition metal compounds.

Electrical properties, metal-insulator transition and high-temperature superconductivity have been studied in the generalized Hubbard model in papers [24]-[34]. The result of these papers is the generalized Hubbard model has much better physics than the Hubbard model, firstly, and usage of the electron-hole asymmetry concept allows one to interpret the peculiarities of physical properties of strongly correlated electron systems, which are not explained by the Hubbard model, secondly. In particular, the experimentally observed electron-hole asymmetries of metal oxides conductivity, of cohesive energy of transition 3d-metals and of superconducting properties of high-temperature superconductors, mentioned above, have been explained within the generalized Hubbard model with correlated hopping in papers [24][25][26][27][28], respectively.

An important puzzle remaining beyond consideration in the cited papers is the question about location of the model chemical potential and its dependence on electron concentration and temperature. Theoretical calculations of
chemical potential dependences can be compared with the experimental data obtained by photoemission spectroscopy methods. Also these dependences of chemical potential could give indication for the existence of phase separation in system upon doping.

Even in the framework of the Hubbard model this problem is still open, in particular the question about the jump in chemical potential of the Hubbard model at half-filling in going from electron to hole doping is under discussion in recent literature (see, for instance, review [8]). On the one hand, in majority of papers it has been found that the jump in chemical potential at half-filling is equal to width of the energy gap. On the other hand, some authors suggest that the discontinuity in chemical potential is strictly smaller than the energy gap as a result of inducing states inside the Mott-Hubbard gap (so called “midgap states”) upon doping the Mott insulators. The first scenario is confirmed by the exact solution [37] of the Hubbard model in one dimension. Also numerical studies of the two-dimensional Hubbard model performed in Ref. [38] by means of the Monte Carlo methods show that when the electron concentration \( n = 1 - \delta \) (\( \delta \) is infinitesimal doping), the chemical potential is located at the top of the lower Hubbard band; when the electron concentration \( n = 1 + \delta \), the chemical potential jumps to the bottom of the upper Hubbard band. Correspondingly, upon electron doping the chemical potential jumps from the top of the lower Hubbard band to the bottom of the upper Hubbard band according to the first scenario. The results of papers [39,40] obtained by means of the Kotliar-Ruckenstein slave bosons [11] for the three dimensional Hubbard model and the quantum Monte Carlo method in infinite dimensions [12] suggest that the jump in chemical potential at half-filling is equal to width of the energy gap.

Using the projective self-consistent technique [13] in infinite dimensions, the authors of papers [4] found that, for any value of \( U \) larger than \( U_c \), doping does induce states inside the Mott-Hubbard gap; the splitting of these states from the edge of the Hubbard band is a fraction of the bare kinetic energy, and remains finite even in the limit where \( U \) is infinite. Therefore the jump in chemical potential for infinitesimal doping is strictly less than the energy gap, so the second scenario described above holds. These conclusions were also supported by an extension [44,45] of the iterated perturbative theory approximation [3] for the Hubbard model away from half-filling. Midgap states have also been predicted [47] to occur upon doping in the infinite dimensional Hubbard model using an analytic variant of the Lanczos continued fraction method [15].

In consequence of the absence of a rigorous result on the described scenarios adequacy the subject is still open. Moreover, even in infinite dimensions this problem is under discussion: different approximations, for example the quantum Monte Carlo method and the iterated perturbative theory, lead to the contrary results. Another question is how correlated hopping does influence on chemical potential. Therefore, the investigation of chemical potential behaviour in the doped Hubbard model as well as in the generalized Hubbard model with correlated hopping is a relevant and important task, which the present paper is devoted.

The paper has the following structure. In Section 2 we formulate Hamiltonian of the generalized Hubbard model with correlated hopping, the quasiparticle energy spectrum of the model is derived by means of a generalized mean-field approximation (an analogue of the projection operation) in the Green function technique. The general properties of the spectrum, and the approximation used by us are discussed. In Sections 3-4 the dependences of chemical potential on the model energy parameters, correlated hopping, temperature and electron concentration are studied. The obtained results are compared with the experimental data on Fermi energy behaviour of real strongly correlated electron systems. Finally, Section 5 is devoted to the conclusions from the obtained results.

II. HAMILTONIAN AND QUASIPARTICLE ENERGY SPECTRUM OF THE MODEL

We start from the natural generalization of the Hubbard model including the matrix elements of electron-electron interaction [3,9]

\[
J(ijk) = \int \int \varphi^*(\mathbf{r} - \mathbf{R}_i) \varphi(\mathbf{r} - \mathbf{R}_j) \frac{\varphi^2(\mathbf{r}' - \mathbf{R}_k)}{|\mathbf{r} - \mathbf{r}'|} |\varphi(\mathbf{r}' - \mathbf{R}_k)|^2 d\mathbf{r} d\mathbf{r}',
\]

which correlate the electron hopping from one lattice site to another (\( \varphi(\mathbf{r} - \mathbf{R}_i) \) is the Wannier function). The Hamiltonian of such a generalized Hubbard model with correlated hopping reads as [2,4,10]

\[
H = H_0 + H_1 + H_1',
\]

\[
H_0 = -\mu \sum_i \left( X_i^\uparrow + X_i^\downarrow + 2X_i^2 \right) + U \sum_i X_i^2,
\]

\[
H_1 = t(n) \sum_{ij\sigma} X_i^{\sigma0} X_j^{0\sigma} + \tilde{t}(n) \sum_{ij\sigma} X_i^{2\sigma} X_j^{\sigma2},
\]

\[
H_1' = \sum_{ij\sigma} X_i^{\sigma0} X_j^{0\sigma} + \tilde{t}(n) \sum_{ij\sigma} X_i^{2\sigma} X_j^{\sigma2},
\]

\[
J(ijk) = \int \int \varphi^*(\mathbf{r} - \mathbf{R}_i) \varphi(\mathbf{r} - \mathbf{R}_j) \frac{\varphi^2(\mathbf{r}' - \mathbf{R}_k)}{|\mathbf{r} - \mathbf{r}'|} |\varphi(\mathbf{r}' - \mathbf{R}_k)|^2 d\mathbf{r} d\mathbf{r}',
\]
\[ H'_1 = t'(n) \sum_{i,j} \left( \eta_\sigma X_i^{\sigma 0} X_j^{\sigma 2} + h.c. \right), \tag{2.5} \]

where \( \mu \) is the chemical potential, the primes at the sums signify that \( i \neq j \), \( X_{ki}^{kl} = |k\rangle \langle l| \) is the Hubbard operator [49], \( X_i^{kl} = X_i^{kl} X_i^{lk} \) is the operator of the number of \( |k\rangle \)-states on \( i \)-site; \( |0\rangle \) denotes the state of site, which is not occupied by an electron (hole), \( |\sigma\rangle \) denotes the state of singly occupied (by an electron with spin \( \sigma \) ) \( i \)-site, \( |2\rangle \) denotes the state of doubly occupied (by two electrons with the opposite spins) \( i \)-site (doublon), \( \eta_\uparrow = -1, \eta_\downarrow = 1 \).

\[ t(n) = t_0 + n \sum_{k \neq j \atop k \neq j} J(ik;jk) = t_0 + nT_1, \tag{2.6} \]

\[ t'(n) = t(n) + 2T_2, \quad t'(n) = t(n) + T_2, \quad T_2 = J(iiij) = J(iiij), \tag{2.7} \]

where \( t_0 \) is the uncorrelated hopping integral (the matrix element of electron-ion interaction), \( n = N_e/N \) is the electron concentration (\( N_e \) is the number of electrons within conduction band, \( N \) is the number of lattice sites).

\( H_0 \) describes the atomic limit of narrow-band models. \( H_1 \) describes the translational hopping of holes and doublons; within the present model the hopping integrals of holes \( t(n) \) and doublons \( t'(n) \) are different (in contrast with narrow-band models of the Hubbard type), this leads to the electron-hole asymmetry mentioned above. \( H'_1 \) describes the processes of paired creation and destruction of holes and doublons.

In the model described by Hamiltonian (2.2) an electron hopping from one site to another is correlated 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 (see expression (2.4)). The peculiarity of model (2.2) is taking into consideration of the correlated hopping \( T_1 \) which cause the concentration dependence of the hopping integrals in contrast with similar generalized Hubbard models.

For half-filling case we introduce the following notations: \( t(n) \equiv t = t_0 + T_1, \quad t'(n) \equiv t = t + 2T_2, \quad t'(n) \equiv t' = t + T_2 \). In this case for \( t' = 0 \) some exact results have been found [21,22,23]. In a simple cubic lattice with coordination number \( z \) metal-insulator transition occurs at

\[ U_c = 2z |t_0|. \tag{2.8} \]

If \( U > U_c \) the ground state of system is a paramagnetic Mott-Hubbard insulator with the concentration of doubly occupied sites \( d = 0 \), the ground state energy is equal to zero.

The single-particle Green function in terms of the Hubbard operators reads as

\[ \langle \langle a_{p\sigma} a_{p'\sigma}^+ \rangle \rangle = \langle \langle X_{p\sigma}^{0\sigma} | X_{p'\sigma}^{0\sigma} \rangle \rangle + \eta_\sigma \langle \langle X_{p\sigma}^{0\sigma} | X_{p'\sigma}^{2\sigma} \rangle \rangle + \eta_\sigma \langle \langle X_{p\sigma}^{2\sigma} | X_{p'\sigma}^{0\sigma} \rangle \rangle + \langle \langle X_{p\sigma}^{2\sigma} | X_{p'\sigma}^{2\sigma} \rangle \rangle. \tag{2.9} \]

The Green function \( \langle \langle X_{p\sigma}^{0\sigma} | X_{p'\sigma}^{0\sigma} \rangle \rangle \) is given by the equation

\[ (E + \mu) \langle \langle X_{p\sigma}^{0\sigma} | X_{p'\sigma}^{0\sigma} \rangle \rangle = \frac{\delta_{pp'\sigma}}{2\pi} \langle X_{p\sigma}^{0\sigma} + X_{p'\sigma}^{0\sigma} \rangle + \langle \langle [X_{0\sigma}^{0\sigma}, H_1] | X_{p'\sigma}^{0\sigma} \rangle \rangle \]
\[ + \langle \langle X_{0\sigma}^{0\sigma}, H'_1 \rangle \rangle | X_{p'\sigma}^{0\sigma} \rangle \rangle. \tag{2.10} \]

with \([A,B] = AB - BA\). Using a variant [24,27] of the generalized mean-field approximation [50] we suppose in Eq. (2.10) that

\[ [X_{p\sigma}^{0\sigma}, H_1] \approx \sum_j \epsilon(pj) X_{j\sigma}^{0\sigma}, \quad [X_{p\sigma}^{0\sigma}, H'_1] \approx \sum_j \epsilon_1(pj) X_{j\sigma}^{2\sigma}, \tag{2.11} \]

where \( \epsilon(pj) \) and \( \epsilon_1(pj) \) are the non-operator expressions. The procedure of \( \epsilon(pj) \) and \( \epsilon_1(pj) \) calculation is described in Ref. [24,27] (here there is a partial equivalence with the slave boson method [11]).

Thus we obtain the closed system of equations for the Green functions \( \langle \langle X_{p\sigma}^{0\sigma} | X_{p'\sigma}^{0\sigma} \rangle \rangle \) and \( \langle \langle X_{0\sigma}^{2\sigma} | X_{p'\sigma}^{0\sigma} \rangle \rangle \). An analogous procedure is realized also in the equations for the other Green functions (2.3).

In this way, we find single-particle Green function (2.9) and quasiparticle energy spectrum. For paramagnetic case in \( k \)-representation the spectrum is [27]:

\[ E_{1,2}(k) = -\mu + \frac{U}{2} + \frac{\epsilon(k) + \bar{\epsilon}(k)}{2} \mp \frac{1}{2} Q(k), \tag{2.12} \]

\[ Q(k) = \sqrt{[\epsilon(k) - \bar{\epsilon}(k) - U]^2 + 4\epsilon_1(k)\bar{\epsilon}(k)}. \tag{2.13} \]
The Fourier components of the quantities defining quasiparticle energy spectrum (2.12) are given by the formulae:

\[ \epsilon(k) = \alpha t_k(n), \quad \tilde{\epsilon}(k) = \tilde{\alpha} \tilde{t}(n), \quad \epsilon_1(k) = \alpha_1 t'_k(n), \quad \epsilon_2(k) = \alpha_2 t'_k(n), \]  

(2.14)

\[ \alpha = -n + 2d + \frac{2(1-d)^2}{2 - n} - \frac{2d(d - n + 1) \tilde{t}(n)}{2 - n} \tilde{t}(n), \]  

(2.15)

\[ \tilde{\alpha} = n - 2d + \frac{2d^2}{n} - \frac{2d(d - n + 1) t(n)}{n} t(n), \]  

(2.16)

\[ \alpha_1 = n - 1 - \frac{2d}{n}, \quad \alpha_2 = -1 - n + \frac{2(1-d)}{2 - n}, \]  

(2.17)

with \( d \) being the concentration of doubly occupied sites.

For \( n < 1 \) and \( U \to \infty \) we obtain

\[ E_1(k) = -\mu + \left( \frac{2}{2 - n} - n \right) t_k(n) \]  

(2.18)

(the lower Hubbard band); if \( n > 1 \) and only the upper Hubbard band is important, we obtain

\[ E_2(k) = -\mu + U + \left( \frac{2}{n} - 2 + n \right) \tilde{t}_k(n). \]  

(2.19)

For the Hubbard model \( (t(n) = \tilde{t}(n) = t'(n) = t_0) \) \( E_1(k) \) for \( n \to 0 \), and \( E_2(k) \) for \( n \to 2 \) get the band form. Moreover, expressions (2.12) describe the exact atomic limit for \( t_0 = 0 \) and the band situation for \( U = 0 \).

At half-filling quasiparticle energy spectrum (2.12) is written in the form (20):

\[ E_{1,2}(k) = -\mu + \frac{(1-2d)(t_k + \tilde{t}_k) + U}{2} \pm \frac{1}{2} F_k, \]  

(2.20)

\[ F_k = \sqrt{[B(t_k - \tilde{t}_k) - U]^2 + (4d t'_k)^2}, \quad B = 1 - 2d + 4d^2, \]  

(2.21)

where the doublon concentration \( d \) is found from the equation

\[ d = \langle X^2 \rangle = \frac{1}{2N} \sum_k \left( \frac{A_k}{\exp \frac{E_k(k)}{\theta} + 1} + \frac{B_k}{\exp \frac{E_k(k)}{\theta} + 1} \right), \]  

(2.22)

with

\[ A_k = \frac{1}{2} - \frac{B(\tilde{t}_k - t_k)}{2F_k} - \frac{U}{2F_k}, \quad B_k = \frac{1}{2} + \frac{B(\tilde{t}_k - t_k)}{2F_k} + \frac{U}{2F_k}, \]  

(2.23)

\( \theta = k_B T \), \( k_B \) is the Boltzmann’s constant, \( T \) is the temperature. For the special case \( t' = 0 \) of the model quasiparticle energy spectrum (2.20) reproduces (see Ref. 20) exact result (2.8).

As a result of the absence of a natural small expansion parameter at intermediate to strong Coulomb interactions in the Hubbard model and its generalizations (this situation is under consideration in the present paper), one does not know a rigorously justified nonperturbative approach in the theory of strongly correlated electron systems. In such a case the methods of mean-field type are useful [3,4], we have used one of these methods. The justification of an approximation is the reproduction of limiting cases and known exact results, and also the accordance with experimental data. Discussing the generalized mean-field approximation used by us we note the following facts.

The approximation reproduces the exact atomic and band limits in the Hubbard model and also describes metal-insulator transition as mentioned above. For the Hubbard model the criterion of metal-insulator transition obtained by means of this approximation is \( U_c = 2w_0 \) \( (w_0 = z \theta_0) \) in agreement with the Mott’s criterion [51] and the result of the random dispersion approximation [3] in infinite dimensions. In the limit of strong interactions the approach satisfies another requirement of a good approximate theory for the Hubbard model at half-filling formulated in Ref. [4]: \( W_{l,u} = 2w_0 \) \( (W_l, W_u \) are the widths of the lower and upper Hubbard bands). For the special case \( t' = 0 \) of the model (2.2) the generalized mean-field approximation reproduces exact criterion (2.8) of metal-insulator transition, the ground state energy and doublon concentration. For very small (large) values of electron concentration and \( U \to \infty \) the approximation gives the band description according to the general physical ideas. The approach
allows also to describe the observed transition in some Mott-Hubbard compounds from a metallic state to an insulating phase with the increase of temperature in a paramagnetic state (see, for example, book, p. 178, 195).

On the other hand, a deficiency of the generalized mean-field approximation is the breakdown of Fermi liquid behaviour of the systems with weak interactions and half-filled band (however, it should be noted that in the present paper we consider the cases of strong and intermediate interactions). The question about Fermi liquid behaviour (i.e. the fulfilment of Luttinger’s theorem) at intermediate to strong interactions in three dimensions is under discussion. This is caused by the reason that Luttinger’s theorem is based on the perturbation expansion, assuming an adiabatic relation between a noninteracting system and an interacting one, which may not be fulfilled for the case of intermediate and strong interactions. In particular, the authors have found the violation of Luttinger’s theorem in the doped Hubbard model, and for the case of intermediate and strong interactions. Note also, that a modification allows to take into account effects caused by the effective interaction of local magnetic moments; at the same time, the main features of quasiparticle energy spectrum still persist.

Another limitation of the generalized mean-field theory is that for small values $t'/U$ the approximaton does not reproduce the physics of the interaction of local magnetic moments as this holds in the framework of the effective Hamiltonian method. Nevertheless, we consider the paramagnetic phase, and in this case the effective inter-atomic exchange interaction (kinematic superexchange) leads to the renormalization of chemical potential only (in the molecular field approximation). Therefore, although the system is metallic away from half-filling, it may be non-Fermi liquid at intermediate and strong interactions because of a violation of Luttinger’s theorem.

The noted generalized mean-field approximation has been applied also for study of metal-insulator transition and properties of generalized Hubbard model with correlated hopping, for description of metal-insulator transition in the doubly degenerate Hubbard model and a doubly orbitally degenerate model with correlated hopping.

### III. CHEMICAL POTENTIAL OF THE MODEL AT HALF-FILLING

Consider the important situation of half-filled band $n = 1$. Chemical potential of the model is given by the equation $(\langle X^0_i \rangle = \langle X^1_i \rangle)$:

$$\sum_k \left( \frac{A_k}{\exp \frac{E_1(k)}{\theta} + 1} + \frac{B_k}{\exp \frac{E_2(k)}{\theta} + 1} \right) \cdot \sum_k \left( \frac{B_k}{\exp \frac{-E_1(k)}{\theta} + 1} + \frac{A_k}{\exp \frac{-E_2(k)}{\theta} + 1} \right).$$

From Eq. (3.1) for the rectangular density of states and $T = 0$ we find chemical potential of the generalized Hubbard model with correlated hopping in the region of metal-insulator transition:

$$\mu = \frac{w}{w + \tilde{w}} U \quad (U \leq w + \tilde{w}),$$

$$\mu = \frac{U}{2} + \frac{w - \tilde{w}}{2} \quad (U > w + \tilde{w}),$$

with $w = z|t|$, $\tilde{w} = z|\tilde{t}|$ being the half-widths of the lower and upper Hubbard band respectively. For the Hubbard model formulae (3.2) and (3.3) give the well-known result $\mu = U/2$. At $t' = 0$ chemical potential of the generalized Hubbard model is equal to $\mu = U/2$ beeing a consequence of the electron-hole symmetry which is a characteristic of the model in this case. Note that the value $U_c = w + \tilde{w}$ corresponds to the metal-insulator transition point of the generalized Hubbard model with correlated hopping.

At arbitrary value of temperature from Eq. (3.1) we find the expression for calculating chemical potential:

$$\int_{-\infty}^{w} \left[ \frac{1}{\exp \frac{-E_2(\epsilon)}{\theta} + 1} - \frac{1}{\exp \frac{E_2(\epsilon)}{\theta} + 1} \right] d\epsilon = 0,$$

where $E_1(\epsilon), E_2(\epsilon)$ are obtained from the respective formulæ (2.24) for $E_1(k)$ and $E_2(k)$ substituting $t_k \rightarrow \epsilon$, $\tilde{t}_k \rightarrow \tilde{t}\epsilon$, $t'_k \rightarrow t'\epsilon$. 

\[5\]
Figs. [1][3] show the dependences of chemical potential on the ratio $U/w$ and temperature are plotted, show that chemical potential of the generalized Hubbard model with correlated hopping $\mu > U/2$ and only in the absence of correlated hopping or within high temperature region the chemical potential is $\mu = U/2$. From Eqs. (3.2) and (3.3) one can see that chemical potential of the generalized Hubbard model with correlated hopping becomes dependent on $U$, $w$, $\tilde{w}$. The dependences on these parameters are different in metallic and insulating phases, this leads to a kink at the point of metal-insulator transition (see Fig. [1]); $t' = 0$ (i.e. $\tilde{w} = 0$) and $t' = 0.5t$ correspond to the values of correlated hopping parameters $\tau_1 = T_1/|t_0| = 0$ and $\tau_2 = T_2/|t_0| = 0.5$. Taking into consideration correlated hopping leads to the non-equivalence of the lower and upper Hubbard bands. This changes the value of chemical potential from the point $\mu = U/2$: with the increase of the correlated hopping $T_2$ (i.e. with decreasing $\tilde{w}$) chemical potential moves towards the upper Hubbard band.

With the increase of temperature the kink in the chemical potential curve disappears (Fig. [2]). Similar peculiarity in the evolution of free energy dependence on parameter $U/w$ with temperature was noted by Mott [51].

From Fig. [3] one can see that in the model under consideration within low and room temperature regions chemical potential is essentially dependent not only on the parameters $w$ and $\tilde{w}$, but becomes also dependent on temperature (in contrast to chemical potential of the Hubbard model). Moreover, with the decrease of temperature chemical potential rapidly increases, this dependence is different for the different values of correlated hopping parameters $\tau_1$, $\tau_2$. The found temperature dependence of chemical potential can be explained by the following reasons. At zero temperature in an insulating phase the chemical potential is located at the centre of the energy gap (between the top of the lower Hubbard band and the bottom of the upper Hubbard band), the chemical potential value is larger than $U/2$. As the temperature increases some electrons are thermally activated and the energy levels within the upper (lower) Hubbard band become to be occupied (empty). The width of the lower Hubbard band is larger than the width of the upper Hubbard band, and thus, the density of states in the lower band is smaller than the density of states in the upper band (as a result that the distance between the nearest energy levels within the lower Hubbard band is larger versus the corresponding distance in the upper Hubbard band). Consequently, with increasing temperature chemical potential position moves towards the lower Hubbard band, and chemical potential value decreases.

Within high temperature region in the generalized Hubbard model chemical potential tends to $U/2$ with the increase of temperature; really, for $T \to \infty$ the probabilities of an electron finding within the lower and upper Hubbard bands are equal.

IV. CHEMICAL POTENTIAL OF THE DOPED MOTT-HUBBARD SYSTEMS

Find the dependence of chemical potential $\mu$ of the generalized Hubbard model with correlated hopping on carrier concentration. Chemical potential is given by the equation $(\langle X_i^2 \rangle - \langle X_i^0 \rangle = n - 1)$:

$$\frac{n}{2N} \sum_k \left( \frac{C_k}{\exp \left( \frac{E_1(k)}{\theta} \right) + 1} + \frac{D_k}{\exp \left( \frac{E_2(k)}{\theta} \right) + 1} \right) - \frac{1 - n/2}{N} \sum_k \left( \frac{D_k}{\exp \left( - \frac{E_1(k)}{\theta} \right) + 1} + \frac{C_k}{\exp \left( - \frac{E_2(k)}{\theta} \right) + 1} \right) = n - 1,$$

(4.1)

with $E_1(k)$ and $E_2(k)$ determining by expression (2.12),

$$C_k = \frac{1}{2} - \hat{\epsilon}(k) - \epsilon(k) + U \frac{Q(k)}{2Q(k)}, \quad D_k = \frac{1}{2} + \hat{\epsilon}(k) - \epsilon(k) + U \frac{Q(k)}{2Q(k)},$$

(4.2)

$Q(k)$ is defined by formula (2.13), and $\hat{\epsilon}(k)$, $\epsilon(k)$ are given by expressions (2.14).

Eq. (4.1) at $T = 0$ is rewritten in the form:

$$\frac{1}{N} \sum_k [D_k(n - 1) + 1 - n/2] \theta(E_2(k)) + \frac{1}{N} \sum_k [D_k(1 - n) + 1/2] \theta(E_1(k)) = 1 - n/2.$$

(4.3)

We assume the rectangular density of states and pass in Eq. (4.3) from a summation over $k$ to an integration over energy. We solve this equation for the case of a paramagnetic phase of the doped Mott-Hubbard insulator (the energy
defined by the quantity with electron concentration dependence is in qualitative agreement with the results of Refs. [59, 60], obtained by means of the Gutzwiller variational model has the linear dependence on correlated hopping parameter \( s \), and this dependence on chemical potential decreases with increasing correlated hopping \( \tau \).

Fig. 4, however, there is another essential difference between them (see the next paragraph). It should be also noted that for these or close values of correlated hopping parameters within the model under consideration the value of phase separation upon doping changes essentially the position of chemical potential of the generalized Hubbard model on the correlated hopping changes essentially the position of chemical potential of the generalized Hubbard model: at \( n < n_1 \) the chemical potential of the generalized Hubbard model is larger than chemical potential of the Hubbard model, and vice versa for the electron concentration \( n > \tau_2 \). For the Hubbard model this dependence is strictly equal to the energy gap width \( \Delta E = U - w - \tilde{w} \). Therefore, our calculations confirm the first scenario of the chemical potential evolution upon doping mentioned in Section 1. As a result of the absence of a rigorous result on the chemical potential behaviour, one of the main tests of an approximate theory is the comparison with experimental data. One knows no experimental data, which show that the theoretical prediction of some authors, such as the existence of mid-gap states in the Mott-Hubbard insulator upon doping, indeed occur in real transition metal compounds. However, the experiment [67, 68] suggests the jump in Fermi energy near half-filling is equal to the Mott-Hubbard gap in some strongly correlated electron systems.

Fig. 4 displays the jump in chemical potential at half-filling in going from electron to hole doping, this discontinuity in chemical potential is strictly equal to the energy gap width \( \Delta E = U - w - \tilde{w} \). Therefore, our calculations confirm the first scenario of the chemical potential evolution upon doping mentioned in Section 1. As a result of the absence of a rigorous result on the chemical potential behaviour, one of the main tests of an approximate theory is the comparison with experimental data. One knows no experimental data, which show that the theoretical prediction of some authors, such as the existence of mid-gap states in the Mott-Hubbard insulator upon doping, indeed occur in real transition metal compounds. However, the experiment [67, 68] suggests the jump in Fermi energy near half-filling is equal to the Mott-Hubbard gap in some strongly correlated electron systems.

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The concentration dependence of the chemical potential \( \mu(n) \) and \( \tilde{w}(n) \) is plotted in Fig. 4. For the Hubbard model this dependence is in qualitative agreement with the results of Refs. [59, 60], obtained by means of the Gutzwiller variational method \([43]\) in three dimensions and the projective self-consistent technique \([33]\) in infinite dimensions. However, the curve in the Fig. 4 is as opposed to that plotted in paper \([52]\): the second derivatives of chemical potential with electron concentration \( n \) are opposite in sign; thus, the behaviour of electron compressibility of system (which is defined by the quantity \( \frac{d^2}{dn^2} \)), determining with the help of Eq. (4.4) and (4.5), is inverse to the compressibility behaviour obtained using \( \mu(n) \)-dependence of Ref. \([33]\). The results of works \([60, 61]\) coincide with the conclusions derived from Fig. 4; however, there is another essential difference between them (see the next paragraph). It should be also noted that for these or close values of correlated hopping parameters within the model under consideration the value of phase separation upon doping changes essentially the position of chemical potential of the generalized Hubbard model: at \( n < n_1 \) the chemical potential of the generalized Hubbard model is larger than chemical potential of the Hubbard model, and vice versa for the electron concentration \( n > \tau_2 \). For the Hubbard model this dependence is strictly equal to the energy gap width \( \Delta E = U - w - \tilde{w} \). Therefore, our calculations confirm the first scenario of the chemical potential evolution upon doping mentioned in Section 1. As a result of the absence of a rigorous result on the chemical potential behaviour, one of the main tests of an approximate theory is the comparison with experimental data. One knows no experimental data, which show that the theoretical prediction of some authors, such as the existence of mid-gap states in the Mott-Hubbard insulator upon doping, indeed occur in real transition metal compounds. However, the experiment [67, 68] suggests the jump in Fermi energy near half-filling is equal to the Mott-Hubbard gap in some strongly correlated electron systems.

Similarly, we can explain the dependence of chemical potential of the generalized Hubbard model on the correlated hopping changes essentially the position of chemical potential of the generalized Hubbard model: at \( n < n_1 \) increasing correlated hopping \( T_1 \) (or \( T_2 \)) leads to the increase of chemical potential, and for \( n > 4/3 \) chemical potential decreases with increasing correlated hopping \( T_1, T_2 \). This can be explained by the following arguments. For the electron concentration \( n \) the bottom of the upper Hubbard band. When the electron concentration is \( n \) the chemical potential position corresponds to the top of the upper Hubbard band. For the electron concentration \( n \) chemical potential of the generalized Hubbard model is larger than chemical potential of the Hubbard model, and vice versa for the electron concentration \( n \) as a result of the fact that width of the upper band of the Hubbard model is larger than width of the upper band of the model with electron-hole asymmetry. Thus, the value \( n_0 \) of electron concentration must exist satisfying the requirements: at this value the chemical potentials of the Hubbard and generalized Hubbard models are equal (in this case the chemical potential is located at the centre of the upper Hubbard band, i.e. \( \mu = U \)), for electron concentration \( n < n_0 \) increasing correlated hopping \( T_1, T_2 \) leads to the increase of chemical potential, and for \( n > n_0 \) chemical potential decreases with increasing correlated hopping \( T_1, T_2 \).

Similarly, we can explain the dependence of chemical potential of the generalized Hubbard model on the correlated hopping \( T_1 \) for electron concentration \( n < 1 \). But in this case, with contrast to the previous situation, the dependence of chemical potential on the correlated hopping \( T_1 \) is weak (besides, for the doped Mott-Hubbard insulators with \( n < 1 \) chemical potential has no dependence on the correlated hopping \( T_2 \)). It should be stressed that the found values \( n_0 = 2/3, 4/3 \) are the electron concentrations corresponding to the change of conductivity type of the system \([24]\).

From Figs. 4, 6 one can see that for electron concentrations \( n > 1 \) the chemical potential of the generalized Hubbard model has the linear dependence on correlated hopping parameters, and this dependence on \( T_2 \) is stronger versus \( T_1 \). For the values of correlated hopping parameters \( T_1 = 0, T_2 = 0.5 \) the chemical potential of the generalized Hubbard model at \( n > 1 \) becomes equal to the chemical potential of the Hubbard model in the atomic limit \( \mu = U \) (though \( t_0 \neq 0 \)); in this case chemical potential of the generalized Hubbard model does not depend on electron concentration. Notice that for these or close values of correlated hopping parameters within the model under consideration the following situation, which is analogous one observed in heavy fermion substances and compounds with mixed valency, can occur. Near the Fermi level which lies within the lower Hubbard band, the level or very narrow upper Hubbard band (being analogous to the f-level in heavy fermion substances and compounds with mixed valency) can arise in
conduction electron subsystem, as a result of electron-electron interactions of correlated hopping type. In particular, this can lead to the very strong enhancement of effective mass of carriers \cite{27,28}.

It should be noted that using more realistic expressions for the density of states does not change essentially the behaviour character of the obtained dependences of chemical potential on the model parameters. For example, in the case of semielliptic density of states:

\[
\frac{1}{N} \sum_k \delta(E - t(k)) = \frac{2}{\pi w} \sqrt{1 - \left(\frac{E}{w}\right)^2}
\]  

we obtain the following equation for chemical potential of the generalized Hubbard model with correlated hopping:

\[
\frac{2 - n}{2\pi} \arcsin \left(\frac{2 - 2n + n^2}{2 - n} \frac{\mu}{w(n)}\right) + \frac{2 - 2n + n^2}{\pi} \sqrt{1 - \left(\frac{2 - 2n + n^2}{2 - n}\right)^2} \frac{\mu^2}{w^2(n)} = \frac{3n}{4} - \frac{1}{2} \quad (n < 1),
\]

\[
\frac{n}{2\pi} \arcsin \left(\frac{(\mu - U)n}{2 - 2n + n^2}w(n)\right) + \frac{(\mu - U)n^2}{2\pi(2 - 2n + n^2)w(n)} \sqrt{1 - \left(\frac{(\mu - U)n}{2 - 2n + n^2}w(n)\right)^2} = \frac{3n}{4} - 1 \quad (n > 1).
\]

Analysis of these equations shows that the obtained behaviour peculiarities of chemical potential for the case of the rectangular density of states persist. Moreover, a choice of the semielliptic density of states does not change even the values \(n_0 = 2/3, 4/3\).

### A. Temperature dependence of chemical potential

Find the dependence of chemical potential of the generalized Hubbard model with correlated hopping upon temperature. Using the rectangular density of states, we rewrite Eq. (4.1) for chemical potential in the form:

\[
\frac{n}{4\tilde{w}(n)} \int_{-\tilde{w}(n)}^{\tilde{w}(n)} \frac{de}{\exp \frac{E_2(\epsilon)}{\theta} + 1} - \frac{1 - n/2}{2w(n)} \int_{-w(n)}^{w(n)} \frac{de}{\exp \frac{E_1(\epsilon)}{\theta} + 1} = n - 1. \quad (4.9)
\]

After integration for the case of the doped Mott-Hubbard insulator we obtain the following equation for chemical potential:

\[
\frac{1}{4\tilde{w}(n)} \ln \frac{1 + \exp \frac{-\mu + U - n\tilde{w}(n)}{\theta}}{1 + \exp \frac{-\mu + U + n\tilde{w}(n)}{\theta}} = \frac{n/2 - 1}{2\beta\tilde{w}(n)} \ln \frac{1 + \exp \frac{\mu + \beta\tilde{w}(n)}{\theta}}{1 + \exp \frac{\mu - \beta\tilde{w}(n)}{\theta}} \quad (n < 1),
\]

\[
\frac{1}{4\tilde{w}(n)} \ln \frac{1 + \exp \frac{-\mu + U - \tilde{w}(n)}{\theta}}{1 + \exp \frac{-\mu + U + \tilde{w}(n)}{\theta}} = -\frac{1}{4w(n)} \ln \frac{1 + \exp \frac{\mu + (2 - n)\tilde{w}(n)}{\theta}}{1 + \exp \frac{\mu - (2 - n)\tilde{w}(n)}{\theta}} \quad (n > 1),
\]

with the coefficients \(\beta = \frac{2 - 2n + n^2}{2 - n}\), \(\tilde{\beta} = \frac{2 - 2n + n^2}{2 - 2n + n^2}\).

On the base of expressions (4.10) and (4.11) we obtain the dependence of chemical potential on electron concentration at some values of temperature (Fig. 3), and \(\mu(T)\)-dependence for some values of electron concentration (Fig. 4). Here we limit ourselves to a consideration of the Hubbard model case, because taking into account correlated hopping of electrons at \(T \neq 0\) does not lead to qualitatively new distinctions of the chemical potential behaviour from the mentioned one at \(T = 0\).

From Fig. 3 one can see that at temperature \(T > 0\) the discontinuous jump in chemical potential, obtained in the previous subsection, becomes more smooth, the dependence of chemical potential on electron concentration becomes a S-shaped curve with flex point at the electron concentration \(n = 1\).

Figs. 3-5 show that for the electron concentration \(n < n_0\) chemical potential of the doped Mott-Hubbard insulator decreases with the increase of temperature, and for \(n > n_0\) the increase of temperature leads to the increase of
chemical potential; at \( n = n_0 \) the chemical potential does not depend on temperature. These peculiarities of the temperature-dependence of chemical potential can be explained by the following reasons. Let us consider the case \( n < 1 \). For small values of electron concentration \( n < n_0 \) the Fermi energy lies below the centre of the lower Hubbard band, thus with the increase of temperature electrons become to occupy the above-laid empty energy levels, this leads to decreasing chemical potential (an analogue of the case of free electrons). For the electron concentration \( n = n_0 \) the Fermi energy is located at the centre of the lower Hubbard band, as the temperature increases the above-laid empty energy levels within the lower Hubbard band become to be occupied, but the chemical potential does not change its value as a result of symmetry about the centre of the band. For enough high temperatures and values of the intra-atomic Coulomb repulsion \( U \geq 2w_0 \) some electrons are thermally activated into the upper Hubbard band, therefore the chemical potential value decreases with increasing temperature at \( n = n_0 \). For values of electron concentration \( n > n_0 \) the number of empty energy levels within the lower Hubbard band is small because the Fermi energy in this case lies above the centre of the lower band, consequently with the increase of temperature chemical potential increases owing to occupancy of these levels and a presence of the empty upper Hubbard band. Similarly, the influence of temperature and electron concentration on chemical potential of the Hubbard model for the values of electron concentration \( n > 1 \) can be explained.

Note that one can see an analogy of the temperature dependence of chemical potential of the model under consideration with the same dependence in doped semiconductors. Really, for electron concentration \( n < n_0 \) within the model the system has \( n \)-type of conductivity \([24]\) and chemical potential decreases with increasing temperature as in \( n \)-type semiconductors. For electron concentration \( n > n_0 \) the system has \( p \)-type of conductivity, correspondingly chemical potential increases with the increase of temperature, as it is characteristically for \( p \)-type semiconductors.

V. CONCLUSIONS

In the present paper chemical potential of the generalized Hubbard model with correlated hopping has been studied by means of a generalized mean-field approximation. The peculiarity of the model in comparison with similar generalized Hubbard models is the concentration dependence of hopping integrals caused by taking into consideration the matrix elements of electron-electron interaction which describe inter-site hopping of electrons (correlated hopping).

The chemical potential as a function of the model energy parameters \( (U, \tilde{t}(n), \tilde{t}(n), \tilde{t'}(n)) \), electron concentration and temperature has been found. At half-filling, in contrast with the Hubbard model where \( \mu = U/2 \), the dependences of chemical potential on energy parameters of the model are different in metallic and insulating phases leading to a kink at the point of metal-insulator transition at zero temperature; with the increase of temperature the kink in the chemical potential curve disappears. With the increase of the correlated hopping \( T_2 \) (i.e. with decreasing \( \tilde{w} \)) chemical potential moves towards the upper Hubbard band.

Chemical potential of the generalized Hubbard model with correlated hopping is temperature-dependent even in the case of half-filled band (in contrast to the Hubbard model). With the decrease of temperature chemical potential rapidly increases, this dependence is different for the different values of correlated hopping parameters \( \tau_1, \tau_2 \). In high temperature region chemical potential of the asymmetric Hubbard model tends to \( U/2 \) as temperature increases.

In strong coupling regime we have found that correlated hopping changes essentially the location of chemical potential of the generalized Hubbard model: for the electron concentration \( n < n_0 \) increasing correlated hopping parameters leads to the increase of chemical potential, and for the case \( n > n_0 \) chemical potential decreases with increasing correlated hopping parameters. Similarly, for the electron concentration \( n < n_0 \) chemical potential decreases with the increase of temperature, and for \( n > n_0 \) the increase of temperature leads to the increase of chemical potential; at the value of electron concentration \( n = n_0 \) the chemical potential does not depend on correlated hopping and temperature. The found values \( n_0 = 2/3, 4/3 \) correspond to the electron concentration when conductivity type of the system changes; chemical potential of the systems with these electron concentrations is located at the centre of the lower and upper Hubbard band respectively.

At zero temperature we have found the jump in chemical potential at half-filling in going from electron to hole doping, this discontinuity in chemical potential is strictly equal to the energy gap width \( \Delta E = U - w - \tilde{w} \). If the temperature is greater than zero then the discontinuous jump of chemical potential becomes more smooth, the dependence of chemical potential on electron concentration becomes a S-shaped curve with flex point at the electron concentration \( n = 1 \).
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FIG. 1. Dependence of chemical potential $\mu$ of the generalized Hubbard model with correlated hopping in the ground state: the upper curve corresponds to $\tau_1 = 0$, $\tau_2 = 0.5$; the middle curve $- \tau_1 = 0$, $\tau_2 = 0.3$; the lower curve $- \tau_1 = \tau_2 = 0$ (the Hubbard model). The asterisks denote the point of metal-insulator transition.

FIG. 2. Evolution of the dependence of chemical potential $\mu$ of the generalized Hubbard model ($\tau_1 = 0$, $\tau_2 = 0.5$) on $U/w$ with temperature $\theta/w = k_B T/w = 0, 0.1, 0.3, 1$ respectively. The lowermost curve corresponds to the behaviour of chemical potential of the Hubbard model.

FIG. 3. Temperature dependence of chemical potential $\mu$ of the generalized Hubbard model with correlated hopping at $U/2w = 1$: the upper curve corresponds to $\tau_1 = \tau_2 = 0.3$; the lower curve $- \tau_1 = \tau_2 = 0.1$; the straight $- \tau_1 = \tau_2 = 0$ (the Hubbard model).

FIG. 4. Chemical potential of the generalized Hubbard model versus the electron concentration $n$ at $U/w_0 = 4$: $\tau_1 = \tau_2 = 0$ (1); $\tau_1 = \tau_2 = 0.2$ (2); $\tau_1 = 0$, $\tau_2 = 0.5$ (3).
FIG. 5. Evolution of the concentration dependence of chemical potential of the Hubbard model with temperature for $U/w_0 = 4$: $\theta/w_0 = k_B T/w_0 = 0, 0.1, 0.3, 0.5$ respectively.

FIG. 6. Chemical potential of the generalized Hubbard model as a function of the correlated hopping parameter $\tau_1 = T_1/|t_0|$ for $U/w_0 = 4$: the lower curve corresponds to $n = 1.7$, the middle $n = 4/3$, the upper $n = 1.2$.

FIG. 7. Chemical potential of the generalized Hubbard model as a function of the correlated hopping parameter $\tau_2 = T_2/|t_0|$ for $U/w_0 = 4$: the lower curve corresponds to $n = 1.7$, the middle $n = 4/3$, the upper $n = 1.2$.

FIG. 8. Temperature dependence of the chemical potential of the Hubbard model with $U/w_0 = 8$ for different values of electron concentration: $n = 0.98, 0.9, 2/3, 0.5, 0.3$ (from top to bottom).