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Energy spectrum of a doubly orbitally degenerate model with non-equivalent subbands

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In the present paper we investigate a doubly orbitally degenerate narrow-band model with correlated hopping. The model peculiarity takes into account the matrix element of electron-electron interaction which describes intersite hoppings of electrons. In particular, this leads to the concentration dependence of the effective hopping integral. The cases of the strong and weak Hund's coupling are considered. By means of a generalized mean-field approximation the single-particle Green function and quasiparticle energy spectrum are calculated. Metal-insulator transition is studied in the model at different integer values of the electron concentration. Using the obtained energy spectrum we find criteria of metal-insulator transition.

Key words: *narrow energy bands, orbital degeneracy, metal-insulator transition, correlated hopping*

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Both theoretical analysis [1–3] and available experimental data [4] point out that the Hubbard model [5] should be generalized by taking into account orbital degeneration and correlated hopping. In the present paper we study a metal-insulator transition in the recently proposed [6] doubly orbitally degenerate narrow-band model with correlated hopping. The peculiarity of the model is the electron-hole asymmetry and the dependence of hopping integral on the average number of electrons per site, thus the model shows much better properties than, for example, the Hubbard model with doubly orbital degeneration. The model Hamiltonian is

$$\begin{aligned} H &= -\mu \sum_{i\gamma\sigma} a^+_{i\gamma\sigma} a_{i\gamma\sigma} + \sum_{ij\gamma\sigma}{}' t_{ij}(n) a^+_{i\gamma\sigma} a_{j\gamma\sigma} + \sum_{ij\gamma\sigma}{}' (t'_{ij} a^+_{i\gamma\sigma} a_{j\gamma\sigma} n_{i\bar{\gamma}} + \text{h.c.}) \\ &\quad + \sum_{ij\gamma\sigma}{}' (t''_{ij} a^+_{i\gamma\sigma} a_{j\gamma\sigma} n_{i\gamma\bar{\sigma}} + \text{h.c.}) + U \sum_{i\gamma} n_{i\gamma\uparrow} n_{i\gamma\downarrow} + U' \sum_{i\sigma} n_{i\alpha\sigma} n_{i\beta\bar{\sigma}} \end{aligned}$$

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$$+(U'-J)\sum_{i\sigma} n_{i\alpha\sigma}n_{i\beta\sigma}, \tag{1}$$

where μ is the chemical potential, $a^{+}_{i\gamma\sigma}, a_{i\gamma\sigma}$ 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 values of orbital states), $n_{i\gamma\sigma} = a^{+}_{i\gamma\sigma}a_{i\gamma\sigma}$ 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} is the 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 the influence of an electron on $\bar{\gamma}$ (γ)-orbital of i- or j-site on hopping process, the prime at the second sum in equation (1) signifies that $i \neq j$, 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 is the intra-atomic exchange interaction energy which stabilizes the Hund's states forming the atomic magnetic moments, and the effective hopping integral $t_{ij}(n) = t_{ij} + nT_1(ij)$ is concentration-dependent due to taking into account the correlated hopping $T_1(ij)$.

The Hamiltonian (1) describes the model with non-equivalent subbands (the analogues of Hubbard subbands). The non-equivalence of the subbands leads to different width of the subbands and different values of the density of states within the subbands. At the same time, the density of states within each subband is symmetrical. As a consequence, the chemical potential is placed between the subbands at integer values of the electron concentration $n = 1,\ 2,\ 3$. In these cases, in the model described by the Hamiltonian (1), the metal-insulator transition (MIT) can occur.

1. Let us consider the case of the strong intra-atomic Coulomb interaction $U' \gg t_{ij}$ and the strong Hund's coupling $U' \gg U' - J$ (values U' and J are of the same order). These conditions allow us to neglect the states of site when there are more than two electrons on the site and the "non-Hund's" doubly occupied states (the analogous conditions are used for an investigation of magnetic properties of the Hubbard model with twofold orbital degeneration in [7–9]). Thus, lattice sites can be in one of the seven possible states: a hole (a non-occupied by electron site); a single occupied by electron site; the Hund's doublon (a site with two electrons on different orbitals with the same spins).

Using the method of works [10–13] we obtain the energy gap (here we neglect the correlated hopping)

$$\begin{aligned} \Delta E &= -2w(0.75 - 1.5c) + (1/2)(F_1 + F_2), \\ F_{1,2} &= \sqrt{[(U'-J) \mp 0.5w)]^2 + 16c^2w^2}, \end{aligned} \tag{2}$$

where $w = z|t|$, z is the number of the nearest neighbours to a site, c is the hole concentration. At $T = 0$ K MIT occurs when $(U'-J)/(2w) = 0.75$.

The energy gap width ΔE as a function of the parameters $(U'-J)/(2w)$ and $(kT)/(2w)$ is presented in figure 1 and figure 2, respectively. With a change of the parameter $(U'-J)/(2w)$ the system undergoes the transition from an insulating to

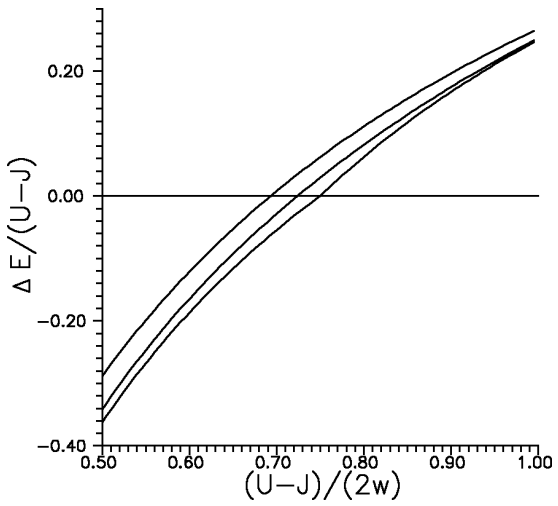

Figure 1. The dependence of energy gap width $\Delta E/(U' - J)$ on the parameter $(U' - J)/(2w)$: the upper curve – $(kT)/(2w) = 0.1$; the middle curve – $(kT)/(2w) = 0.05$; the lower curve – $(kT)/(2w) = 0$.

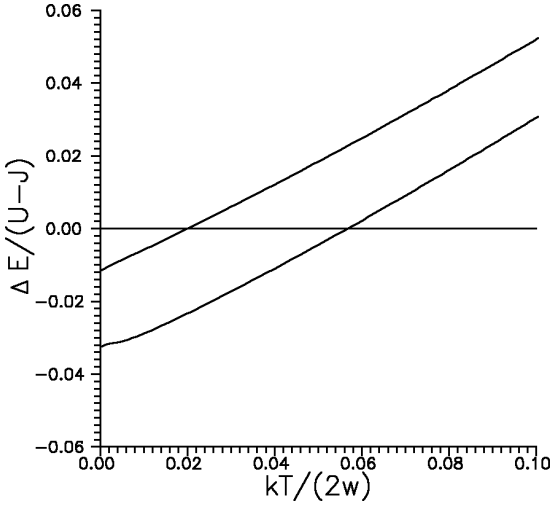

Figure 2. The dependence of energy gap width $\Delta E/(U' - J)$ on the parameter $(kT)/(2w)$: the upper curve – $(U' - J)/(2w) = 0.74$; the lower curve – $(U' - J)/(2w) = 0.72$.

a metallic state (negative values of the energy gap width correspond to the overlapping of the Hubbard subbands). In the model under consideration at $T = 0$ K, an insulator-metal transition at $n = 1$ occurs when $(U' - J)/(2w) = 0.75$ (figure 1, the lower curve).

The transition from a metallic to an insulating state with the increase of temperature at a given value of the parameter $(U' - J)/(2w)$ is also possible (figure 2). It can be explained by the fact that the energy gap width ΔE given by equation (2) increases with the temperature T increase which is caused by the rise of the polar states concentration at constant w, $(U' - J)$.

2. The exchange interaction splits some of the bands. If the exchange interaction is small comparative to the Coulomb interaction $J \ll U$, then the splitting is small and leads only to a weak broadening of the bands. Forasmuch we calculate the width of the energy gap we can take into account the effect of J by an appropriate shift of the band center resulting from the inclusion of J into the chemical potential by means of mean-field approximation (see, e.g., [6,14]).

To describe MIT at the electron concentration n, we can take into account in the Hamiltonian only the states of site with $n - 1,\ n,\ n + 1$ electrons (the analogous simplification has been used in [15,16]). In the vicinity of the transition point at the electron concentration $n = 1$, the concentrations of sites occupied by three and four electrons are small. We can neglect the small amounts of these sites. For calculation of single-particle Green functions we use the generalized mean-field approximation [10]. After transition to **k**-representation, we obtain the quasiparticle

energy spectrum:

$$E_{1,2}(\mathbf{k}) = -\tilde{\mu} + \frac{U}{2} + \frac{\epsilon(\mathbf{k}) + \zeta(\mathbf{k})}{2} \mp \frac{1}{2}\left\{[U - \epsilon(\mathbf{k}) + \zeta(\mathbf{k})]^2 + 4\tilde{\epsilon}(\mathbf{k})\tilde{\zeta}(\mathbf{k})\right\}^{1/2}. \quad (3)$$

By use of the mean-field approximation, in the case of $t'_k = t''_k$ we obtain $\epsilon(\mathbf{k}), \tilde{\epsilon}(\mathbf{k}), \zeta(\mathbf{k}), \tilde{\zeta}(\mathbf{k})$ as functions of $\tilde{t}_{\mathbf{k}} = t_{\mathbf{k}} + 2t'_{\mathbf{k}}$ and $c,\ b,\ d$ being the concentrations of the holes and sites occupied by one, two electrons, respectively, connected by the relations: $c = 6d$, $b = \frac{1}{4} - 3d$. In the transition point, when the concentrations of the holes and doublons are equal to zero, the energies of the electrons within the subbands are

$$\begin{aligned} E_1(\mathbf{k}) &= -\tilde{\mu} + t_{\mathbf{k}}, \\ E_2(\mathbf{k}) &= -\tilde{\mu} + U + \tilde{t}_{\mathbf{k}}. \end{aligned} \quad (4)$$

From the equations (4) we obtain the criterion of MIT: $U = w + \tilde{w}$, where $w = z|t_{ij}|$, $\tilde{w} = z|\tilde{t}_{ij}|$. With the increase of the correlated hopping at the fixed value of parameter $U/2w$, the energy gap width increases and the region of values of $U/2w$ at which the system is in a metallic state, decreases. In the partial case $t'_{\mathbf{k}} = t''_{\mathbf{k}} = 0$ (in this case $t_{\mathbf{k}} = \tilde{t}_{\mathbf{k}}$) we have $U_c/2w = 1$.

Let us consider the MIT at electron concentration $n = 2$. In the vicinity of the transition point in the case of two electrons per atom, the concentrations of holes and sites occupied by four electrons are small. For the small values of the intra-atomic exchange interaction ($J \ll U$) we take J into account analogously to the case of $n = 1$. To calculate single-particle Green functions we use the generalized mean-field approximation. After transition to $\mathbf{k}$-representation, we obtain the quasiparticle energy spectrum:

$$E_{1,2}(\mathbf{k}) = -\tilde{\mu} + \frac{3U}{2} + \frac{\epsilon(\mathbf{k}) + \zeta(\mathbf{k})}{2} \mp \frac{1}{2}\left\{[U - \epsilon(\mathbf{k}) + \zeta(\mathbf{k})]^2 + 4\tilde{\epsilon}(\mathbf{k})\tilde{\zeta}(\mathbf{k})\right\}^{1/2}. \quad (5)$$

By use of the mean-field approximation analogously to the above, in the case of $t'_k = t''_k$ we obtain $\epsilon(\mathbf{k}), \tilde{\epsilon}(\mathbf{k}), \zeta(\mathbf{k}), \tilde{\zeta}(\mathbf{k})$ as functions of $\tilde{t}_{\mathbf{k}} = t_{\mathbf{k}} + 2t'_{\mathbf{k}}$, $t^*_{\mathbf{k}} = t_{\mathbf{k}} + 4t'_{\mathbf{k}}$ and $b,\ d$, where b is the concentration of the sites occupied by one (or three) electrons, d is the concentration of the doubly occupied sites, connected by the relation $b = (1 - 8d)/6$.

In the transition point, when the concentrations of the singly and triply occupied sites are equal to zero, the quasiparticle energy spectrum is

$$E_{1,2}(\mathbf{k}) = -\tilde{\mu} + \frac{3U}{2} + \frac{17}{18}\frac{t^*_{\mathbf{k}} + \tilde{t}_{\mathbf{k}}}{2} \mp \frac{1}{2}\left\{\left[U + \frac{17}{18}\frac{t^*_{\mathbf{k}} - \tilde{t}_{\mathbf{k}}}{2}\right]^2 + \left[\frac{t^*_{\mathbf{k}} + \tilde{t}_{\mathbf{k}}}{18}\right]^2\right\}^{1/2}. \quad (6)$$

Using the quasiparticle energy spectrum (6), we find the energy gap width. In the point of MIT the energy gap is equal to zero. From this condition we find the criterion of MIT. With the increase of the correlated hopping at the fixed value of parameter $U/2w$, the energy gap width increases faster than at $n = 1$ and the region of values of $U/2w$ at which the system is in the metallic state, decreases, analogously

to the case $n = 1$. In the partial case of $t'_{\mathbf{k}} = t''_{\mathbf{k}} = 0$ (in this case $t^*_{\mathbf{k}} = \tilde{t}_{\mathbf{k}}$) we find $U_c/2\omega = 2\sqrt{2}/3$.

In a similar way, we consider the case of electron concentration $n = 3$. In the vicinity of the transition point in the case of three electrons per atom, the concentrations of holes and sites occupied by one electron are small. Neglecting the small amounts of these sites, we can calculate the single-particle Green functions analogously to the above. We find the values of $\epsilon(\mathbf{k}), \tilde{\epsilon}(\mathbf{k}), \zeta(\mathbf{k}), \tilde{\zeta}(\mathbf{k})$ using the mean-field approximation. They are functions of $t^*_{\mathbf{k}} = t_{\mathbf{k}}+4t'_{\mathbf{k}}$, $t^\bullet_{\mathbf{k}} = t_{\mathbf{k}}+6t'_{\mathbf{k}}$ and $d,\ t,\ f$ being the concentrations of the sites occupied by two, three and four electrons, respectively, connected by the relations: $f = 6d,\ t = 1/4 - 3d$.

In the transition point, when the concentrations of the holes and single electrons are equal to zero, the energies of the electrons within the subbands are

$$\begin{aligned} E_1(\mathbf{k}) &= -\tilde{\mu} + 2U + t^*_{\mathbf{k}}, \\ E_2(\mathbf{k}) &= -\tilde{\mu} + 3U + t^\bullet_{\mathbf{k}}. \end{aligned} \tag{7}$$

From the equation (7) we obtain the criterion of the MIT at the electron concentration $n = 3$: $U = w^* + w^\bullet$, where $w^* = z|t^*_{ij}|$, $w^\bullet = z|t^\bullet_{ij}|$. With the increase of the correlated hopping at the fixed value of parameter $U/2w$, the energy gap width increases faster than at $n = 1, n = 2$ and the region of values of $U/2w$ at which the system is in a metallic state, decreases. In the partial case $t'_{\mathbf{k}} = t''_{\mathbf{k}} = 0$ (in this case $t_{\mathbf{k}} = \tilde{t}_{\mathbf{k}}$) we have $U_c/2w = 1$. This result coincides with the corresponding critical value at the electron concentration $n = 1$ due to the electron-hole symmetry of the model without the correlated hopping.

The peculiarities of the expressions for the quasiparticle energy spectrum are the dependences on the concentration of polar states (holes, doublons at $n = 1$; single electron and triple occupied sites at $n = 2$; doublons and sites occupied by four electrons at $n = 3$) and on the hopping integrals (thus on external pressure). At given values of U and hopping integrals (constant external pressure), the concentration dependence of ΔE permits to study MIT under the action of external effects. In particular, $\Delta E(T)$-dependence can lead to the transition from a metallic state to an insulating state with the increase of temperature (see figure 4). The described transition is observed, in particular, in the $(V_{1-x}Cr_x)_2O_3$ compound [4,17] and the $NiS_{2-x}Se_x$ system [18,19]. The similar dependence of the energy gap width can be observed at the change of the polar states concentration under the action of photoeffect or magnetic field. The strong magnetic field can lead, for example, to the decrease of the polar state concentration (see [20]) initiating the transition from a paramagnetic insulator state to a paramagnetic metal state. The increase of the polar state concentration under the action of light, stimulates the metal-insulator transition, analogously to the influence of temperature change. At the increase of bandwidth (for example, under the action of external pressure or composition changes) the insulator-to-metal transition can occur.

If the correlated hopping is absent in the case $n = 2$, the MIT occurs at the smaller value of $U/2w$ than in the case $n = 1$ (figure 3). This result is in qualitative accordance with the results of work [14], in distinction from [16,21]. Using the

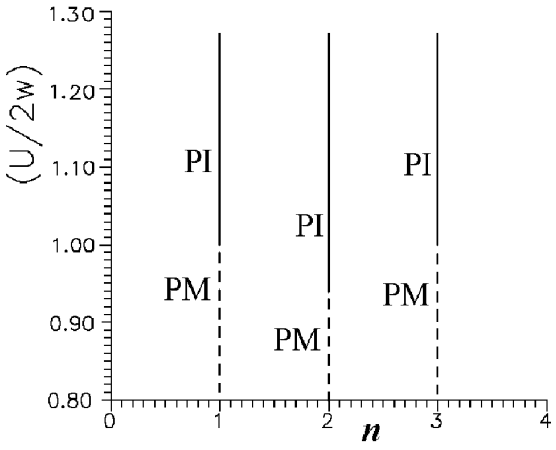

Figure 3. The electron concentration vs. interaction strength phase diagram showing the paramagnetic metal (PM) and paramagnetic insulator (PI) in the absence of correlated hopping.

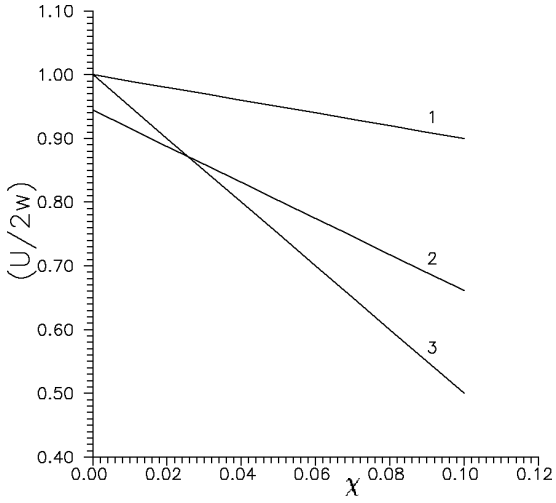

Figure 4. The dependence of critical value $(U/2w)_{\rm c}$ on the parameter of correlated hopping $\chi = t'_{ij}/t_{ij}$: the curve 1 – $n = 1$; the curve 2 – $n = 2$; the curve 3 – $n = 3$.

critical values of the parameter $U/(2w)$ at which MIT occurs for different integer electron concentrations (see figure 3) we can interpret the fact that in the series of disulphides MS_2, the CoS_2 (one electron within e_g band corresponding to $n = 1$) and CuS_2 compounds (three electrons within e_g-band corresponding $n = 3$) are metals, and the NiS_2 compound (two electrons within e_g-band corresponding $n = 2$) is an insulator. Really, for $0.94 \leqslant U/2w \leqslant 1$ at the electron concentration $n = 2$ the system described by the present model is an insulator, whereas for the same values of the parameter $U/2w$ at the electron concentrations $n = 1,\ 3$ the system is a metal (according with the calculations of [22] the ratios $U/2w$ in these compounds have close values).

We have found that in the case of the strong Hund's coupling at $n = 1$, the metal-insulator transition occurs at a smaller value of the parameter $((U - J)/2w)_{\rm c} = 0.75$ than in the case of the weak Hund's coupling $((U - J)/2w)_{\rm c} = 1$.

At nonzero values of correlated hopping, the point of MIT moves towards the values of parameter $U/2w$ at which the system is a metal (figure 4). The non-equivalence of the cases $n = 1$ and $n = 3$ is a manifestation of the electron-hole asymmetry which is a characteristic of the models with correlated hopping.

Thus, both orbital degeneracy and correlated hopping are the factors favouring the transition of the system to an insulating state in the case of half-filling with the increase of intra-atomic Coulomb repulsion in comparison with the single-band Hubbard model.

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Перехід метал-діелектрик у двічі орбітально виродженій моделі з нееквівалентними підзонами

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У роботі ми вивчаємо двократно орбітально вироджену вузькозонну модель з корельованим переносом електронів. Особливістю моделі є врахування матричного елемента електрон-електронної взаємодії, який описує міжвузлові переходи електронів. Це приводить, зокрема, до концентраційної залежності ефективного інтеграла переносу. Розглянуті випадки сильного та слабкого гундівського зв'язку. За допомогою узагальненого наближення середнього поля розраховані одночастинкова функція Гріна та енергетичний спектр. Перехід метал-діелектрик у моделі досліджений при різних цілих значеннях електронної концентрації. За допомогою отриманого енергетичного спектра знайдено критерії переходу метал-діелектрик.

Ключові слова: *вузькі зони провідності, орбітальне виродження, перехід метал-діелектрик, корельований перенос*

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