A microscopical model of doped fulleride electronic subsystem taking the triple orbital degeneracy of energy states into account is considered within the configurational-operator approach. Using the Green function method, the energy spectrum at the integer band filling $n = 1$ corresponding to AC$_{60}$ compounds is calculated. A possible correlation-driven metal-insulator transition within the model is discussed.

1. Introduction

Electrical, optical, and mechanical properties of fullerenes [1, 2] in the condensed state demonstrate a considerable physical content of phenomena, which take place in fullerenes, and show that the use of such materials in electronics has significant perspectives. Fullerene crystals and films are semiconductors with an energy gap of 1.2–1.9 eV [3, 4] and have photoconductivity under visible light irradiation. Fullerene crystals have comparatively small binding energy, and the phase transition connected with the orientational disordering of fullerene molecules occurs in such crystals at room temperature [5].

The addition of radicals containing platinum-group metals [6] to fullerenes C$_{60}$ allows one to obtain ferromagnetic materials based on fullerene. In polycrystal C$_{60}$ doped by an alkali metal, the superconductivity at temperatures lower than 33 K is observed [7, 8]. The large binding energy is typical of metallocarbonohedra M$_8$C$_{12}$, where M = Ti, V, Hg, Zr. For example, in Ti$_8$C$_{12}$, the molecule binding energy per atom is 6.1 eV [9] (for a C$_{60}$ molecule, this energy is 7.4–7.6 eV [3]).

Fullerenes in the solid state (fullerites) are molecular crystals, where the interaction between atoms in C$_{60}$ molecule is much larger than the interaction between nearest molecules. In a closely packed structure, each fullerene molecule has 12 nearest neighbors. Depending on peculiarities of the molecular interaction, the face-centered cubic lattice or hexagonal lattice is realized [10]. A phase transition in the C$_{60}$ crystal occurs at a temperature of 257 K, and this is the first-order transition. At high temperatures, molecules can freely rotate, whereas the rotation at low temperatures is stopped, and the interaction anisotropy of neighbor molecules C$_{60}$ becomes important. This leads to a small sharp change of the distance between the nearest molecules. According to the results of X-ray structure analysis [11], the lattice constant changes from 1.4154 ± 0.0003 nm to 1.4111 ± 0.0003 nm (that is, by 0.43 ± 0.06 percent).

At low temperatures, when C$_{60}$-molecules are oriented in space, the crystal lattice symmetry does not coincide with the symmetry of a single molecule C$_{60}$ (icosahedral symmetry Y). In a unit cell of the fullerite crystal lattice, there are four C$_{60}$-molecules. These molecules form a tetrahedron, in which orientations of all molecules are the same. Tetrahedra, in turn, form a simple cubic lattice.

Fullerites are semiconductors with an energy gap of 1.5–1.95 eV [3]. The electrical resistivity of polycrystals C$_{60}$ [11] monotonically changes with the temperature, and the energy gap has monotonic dependence on the pressure: an increase of the energy gap under the pressure higher than $2 \times 10^5$ atm indicates the absence of the metal-insulator transition at $p \approx 10^6$ atm. In the temperature region 150–400 K, the relaxation time is temperature-independent, which indicates that the carriers are localized, and the hopping mechanism of recombination, which includes the tunneling of electrons between localized states, is realized.

It has been shown in [7] that the doping of solid fullerenes C$_{60}$ by a small amount of an alkali metal leads to the formation of a material with the metallic type of conductivity, and this material becomes superconducting at low temperatures ($T_c$ from 2.5 K for Na$_2$K$_6$C$_{60}$...
to 33 K for RbCs$_2$C$_{60}$). At changes of the temperature, concentration of an alkali metal, and parameters and structure of the lattice, various phases of these compounds have been realized. In particular, at various fillings $n$ ($n$ may change from 0 to 6) of the lowest unoccupied molecular orbital (LUMO), the metallic, insulating, or superconducting phases have been realized. Superconductivity in doped fullerenes K$_x$C$_{60}$ has been studied theoretically in [12], and strong electron correlations have been shown to play a crucial role in the superconducting state stabilization. Recently, the strong electron correlation was also proven [13] to be responsible for the superconductivity of planar carbon systems of the graphene type.

Let us consider the electronic structure of C$_{60}$ in detail. In the single-particle approximation with neglect of electron correlations, the following spectrum has been calculated [2]: 50 of 60 $p_z$ electrons of a neutral molecule fill all orbitals up to $L = 4$. The lowest $L = 0, 1, 2$ orbitals correspond to icosahedral states $a_{3g}, t_{1u}, h_g$. All states with greater $L$ undergo the icosahedral-field splitting. There are 10 electrons in the partially filled $L = 5$ state. The icosahedral splitting ($L = 5 \rightarrow h_u + t_{1u} + t_{2u}$) of this 11-fold degenerate orbital leads to the electronic configuration shown below. Microscopic calculations and experimental data show that the completely filled highest occupied molecular orbital is of $t_{1u}$ symmetry, and LUMO (3-fold degenerate) has $t_{1u}$ symmetry. Under such conditions, the HOMO-LUMO gap appears due to the icosahedral perturbation in the shell with $L = 5$; the energy gap found experimentally is about 1 eV for molecules in vacuum. The $t_{2u}$ (LUMO +1)-state originated from $L = 6$ shell is found approximately to be 1 eV above the $t_{1u}$ LUMO.

Electron-electron correlations in C$_{60}$ are described by two main parameters: the intramolecular Coulomb repulsion $U$ and Hund’s coupling $J_H$. In fullerenes, the competition between the intrasite Coulomb interaction (Hubbard $U$) and delocalization processes related to the translational motion of electrons (which determines the bandwidth) causes the realization of the insulator or metallic state [14]. The majority of experimental data and theoretical calculations indicate that all materials with ions C$_{60}^{n-}$ at integer $n$ are Mott–Hubbard insulators, as $U$ is quite large for all doped compounds A$_x$C$_{60}$. Fullerides A$_x$C$_{60}$ doped with alkali metals A attract much attention of researchers due to the unusual metal-insulator transition in these compounds. Only A$_2$C$_{60}$ is metallic, and other phases AC$_{60}$, A$_2$C$_{60}$ and A$_3$C$_{60}$ are insulator ones [15]. This experimental fact contradicts the results of band structure calculations (see, e.g., [16]), which predict the purely metallic behavior. It has been noted in [17] that, to explain the metallic behavior of the Mott–Hubbard system ($x = 3$ corresponds to the half-filling of the conduction band), one has to consider a degeneracy of the energy band. On the base of the Gutzwiller variational approach, the metal-insulator transition has been proven [17] to exist for all integer band fillings. It is shown that the critical value of Coulomb interaction parameter depends essentially on the band filling and the degeneracy (in the case of half filling, $U_{cr} \approx 2.8$ for double degeneracy, $U_{cr} \approx 3.9$ for triple degeneracy). The present study is devoted to the investigation of the Mott–Hubbard localization in the electronic subsystem of fullerides with strong electron correlations within the model involving the orbital degeneracy of energy levels, strong Coulomb interaction, and correlated hopping of electrons.

2. Hamiltonian of the Doped Fulleride Electronic Subsystem

Within the second quantization formalism, the Hamiltonian of interacting electron systems can be written [18] as

$$H = -\mu \sum_{i\lambda\sigma} a_{i\lambda\sigma}^+ a_{i\lambda\sigma} + \sum_{ij\lambda\sigma} t_{ij} a_{i\lambda\sigma}^+ a_{j\lambda\sigma}$$

$$+ \frac{1}{2} \sum_{ijkl\lambda\rho\gamma\sigma} J^{ijkl\lambda\rho\gamma\sigma} a_{i\lambda\sigma}^+ a_{j\lambda\rho}^+ a_{k\rho\gamma} a_{l\gamma\sigma}, \quad (1)$$

where the first sum with the matrix element

$$t_{ij} = \int d^3r \phi_i^*(r-R_i) \times$$

$$\times \left[ -\frac{\hbar^2}{2m} \Delta + V^{\text{ion}}(r) \right] \phi_j(r-R_j) \quad (2)$$

describes the translational motion (hopping) of electrons in the crystal field $V^{\text{ion}}(r)$, and the second sum is the general expression for pair electron interactions described by the matrix elements

$$J^{ijkl\lambda\rho\gamma\sigma} = \int d^3r \phi_i^*(r-R_i) \phi_j(r-R_j) \times$$

$$\times \frac{e^2}{|r-r'|} \phi_k^*(r-R_k) \phi_l(r-R_l) dr dr', \quad (3)$$

In the above formulae, $a_{i\lambda\sigma}^+$, $a_{i\lambda\sigma}$ are the operators of spin-$\sigma$ electron creation and annihilation in the orbital.
are placed close enough to provide a substantial screen-
lead to the value 1.4-1.6 eV [24, 25] for energy [23]
gives 

Experimental estimation of the electron repulsion
The local density approximation (LDA) gives 3.0 eV [21,

However, it has been shown that these models lack the
possibility to describe the electron-hole asymmetry ob-
erved in real correlated electron systems. To maintain
such a possibility, we are to consider the structure of en-
ergy levels and to estimate interaction parameters prior
to make simplifications. Following works [19,20], we de-
vide the Hamiltonian which includes the correlated hop-
ping of electrons (the site-occupation dependence of hop-
ping parameters results from taking the interactions with
second-order matrix elements into account) and a vari-
ty of intrasite interactions caused by the triple orbital
degeneracy of LUMO in doped fullerenes. The zero-order
interaction integral includes the on-site Coulomb correla-
tion (characterized by the Hubbard parameter U):

\[ U = \int \int |\phi_\lambda^*(\mathbf{r} - \mathbf{R}_i)|^2 \frac{e^2}{|r - r'|} |\phi_\lambda(\mathbf{r}' - \mathbf{R}_i)|^2 d\mathbf{r} d\mathbf{r}'. \]  (4)

In an orbitally degenerate system, the on-site (Hund’s
rule) exchange integral

\[ J_H = \int \int |\phi_\lambda^*(\mathbf{r} - \mathbf{R}_i)| \phi_\lambda'(\mathbf{r} - \mathbf{R}_i) \frac{e^2}{|r - r'|} \times \]

\[ \times |\phi_\lambda(\mathbf{r}' - \mathbf{R}_i)| \phi_\lambda'(\mathbf{r}' - \mathbf{R}_i) d\mathbf{r} d\mathbf{r}', \]  (5)

is of principal importance as well. The parameter U for
fullerenes had been estimated within different methods.
The local density approximation (LDA) gives 3.0 eV [21,
22]. Experimental estimation of the electron repulsion
energy [23] gives \( U \approx 2.7 \) eV.

It is worth to note that, in the solid state, molecules
are placed close enough to provide a substantial screen-
ing of the interaction. The calculation with regard for
the screening effect gives \( U \approx 2.7 \) eV [21, 22]. Combin-
ing Auger spectroscopy and photoemission spectroscopy
lead to the value 1.4-1.6 eV [24, 25] for \( U \). We also
note that the energy cost of electron configurations with
spins aligned in parallel is considerably less than that
for the antiparallel alignment. Orbitally degenerate levels
are filled according to Hund’s rule. Experimental

methods [24] give 0.2 eV ± 0.1 eV for the singlet-triplet
splitting; whereas its value is close to 0.05 eV according
to [26]. The relevant intersite parameters are the elec-
tron hopping integral and the intersite exchange coupling
\( J(\lambda \lambda' j j' \lambda \lambda') \).

The resulting Hamiltonian of the doped fulleride elec-
tronic subsystem reads

\[ H = -\mu \sum_{i} \bar{a}_{i \lambda \sigma}^+ a_{i \lambda \sigma} + U \sum_{\lambda \lambda'} \left( n_{i \lambda \sigma} n_{i \lambda' \sigma} + \frac{U''}{2} \sum_{i \lambda \sigma} n_{i \lambda \sigma} n_{i \lambda' \sigma} + \right) \]

\[ + \sum_{ij \lambda \sigma} t_{ij}(n) a_{i \lambda \sigma}^+ a_{j \lambda \sigma} + \sum_{ij \lambda \sigma} t_{ij}' \left( a_{i \lambda \sigma}^+ a_{j \lambda \sigma} n_{i \lambda} + h.c. \right) + \]

\[ + \sum_{ij \lambda \sigma} t_{ij}' \left( a_{i \lambda \sigma}^+ a_{j \lambda \sigma} n_{i \lambda} + h.c. \right), \]  (6)

where \( n_{i \lambda \sigma} = a_{i \lambda \sigma}^+ a_{i \lambda \sigma} \), \( U'' = U - 2J_H \), and the hop-
ping integrals \( t_{ij}(n), t_{ij}' \), with regard for three types
of correlated hopping of electrons [28] are introduced.

In a model of triply degenerate band, every site can be
in one of 64 configurations (see Fig. 1). To pass from
the electron operator to the Hubbard operators \( X^{pl} \) of
the transition from state \( |l) \) to state \( |p) \), we use the relations

\[ \bar{a}_{\lambda \sigma}^+ = X_{100,000}^{100} + X_{200,000}^{200} + X_{110,010}^{110} + X_{110,001}^{110} + \]

\[ + X_{101,001}^{101} + X_{101,010}^{101} + X_{120,020}^{120} + X_{122,022}^{122} + \]

\[ + X_{122,012}^{122} + X_{122,020}^{122} + X_{122,022}^{122} + \]

\[ + X_{122,122}^{122} + \]

\[ \bar{a}_{\lambda \sigma}^- = X_{100,000}^{100} - X_{200,000}^{200} + X_{110,010}^{110} + X_{110,001}^{110} + \]

\[ + X_{101,001}^{101} + X_{101,010}^{101} + X_{120,020}^{120} + X_{122,022}^{122} + \]

\[ + X_{122,012}^{122} + X_{122,020}^{122} + X_{122,022}^{122} + \]

\[ + X_{122,122}^{122} + \]

\[ \bar{a}_{\lambda \sigma}^{\pm} = X_{100,000}^{100} \pm X_{200,000}^{200} + X_{110,010}^{110} + X_{110,001}^{110} + \]

\[ + X_{101,001}^{101} + X_{101,010}^{101} + X_{120,020}^{120} + X_{122,022}^{122} + \]

\[ + X_{122,012}^{122} + X_{122,020}^{122} + X_{122,022}^{122} + \]

\[ + X_{122,122}^{122} + \]
Fig. 1. Possible site configurations in the threefold degenerate model. The first symbol in the state notation corresponds to the $\alpha$ orbital, the second and third – to the $\beta$ and $\gamma$ orbitals, respectively

$$
\begin{align*}
+X_{101.001}^{101.001} + X_{101.001}^{101.001} + X_{120.020}^{120.020} + X_{102.002}^{102.002} - X_{220.220}^{220.220} + X_{221.021}^{221.021} - X_{222.222}^{222.222}
- X_{220.220}^{220.220} + X_{221.021}^{221.021} + X_{222.222}^{222.222}
- X_{220.220}^{220.220} + X_{221.021}^{221.021} + X_{222.222}^{222.222}
- X_{220.220}^{220.220} + X_{221.021}^{221.021} + X_{222.222}^{222.222}
\end{align*}
$$

which ensure the fulfillment of the anticommutation relations \( \{ X^{\kappa}_{i}, X^{\ell}_{j} \} = \delta_{ij}(\delta_{\kappa\ell}X^{\kappa}_{i} + \delta_{\ell\kappa}X^{\ell}_{j}) \), and the nor-
representation has the form \( \langle p | \rangle \)-state on site \( i \). Such a representation of electronic operators is typical for models of strongly correlated electron systems such as superconducting cuprates [29], manganites [30], cobaltites [31], and optical lattices [32, 33]. Using the root vector notations introduced in [34] allows us to obtain a much more compact form of the Hamiltonian in the configurational representation. In our case, the number of subbands is relatively small, and we use bulky but simple notations which make the projection procedure used below more transparent.

In the configurational representation, the model Hamiltonian takes the form \( H = H_0 + T \). Here, \( H_0 \) sums the “atomic limit” terms, and the translational part can be decomposed as \( T = \sum_{n,m} T_{nm} \), where \( n, m \) serve for numbering the “atomic” states. Terms \( T_{nm} \) of the Hamiltonian form the energy subbands, and terms \( T_{nm} \) describe the hybridization of these subbands. Different hopping integrals correspond to transitions in (or between) the different subbands. The subbands of higher-energy processes appear to be narrower due to the correlated hopping of electrons. The relative positions and the overlapping of subbands depends on the relations between the energy parameters. At the integer values of electron concentration \( \langle n = 1, 2, 3, 4, 5 \rangle \) in the system, the metal-insulator transition is possible. In the partial case of the band filling \( n = 1 \), strong Coulomb correlation, and strong Hund’s coupling (the parameter \( U - 3J_H \) is much greater than the bandwidth, see estimations in [21, 22]), the states with three and more electrons on the same site are excluded. Then the influence of the correlated hopping can be described by three different hopping integrals. The bare band hopping integral \( t_{ij} \) is renormalized with regard for the band narrowing caused by the concentration-dependent correlated hopping as \( t_{ij}(n) = t_{ij}(1 - \tau_1 n) \). This hopping integral characterizes the lower Hubbard subband. The parameter \( \tau_1 \) is usually neglected, but it is of basic importance for a consistent description of correlation effects in narrow band systems (see [19, 20] for a detailed discussion). The hopping integral for the upper Hubbard subband is \( t_{ij}(n) = t_{ij}(n) + 2t'_{ij} \), and \( t_{ij}(n) = t_{ij}(n) + t'_{ij} \) describes a hybridization of the lower and upper Hubbard subbands. In what follows, only the case \( n = 1 \) is considered; so, we omit the explicit notation of concentration dependence. Then the Hamiltonian in the \( X \)-operator representation [27] has the form

\[
H = H_0 + \sum_{\lambda=0,\beta,\gamma} \left( H_b^{(\lambda)} + H_h^{(\lambda)} \right),
\]

where

\[
H_0 = -\mu \sum_{i,\sigma} \left( X_{i}^{\sigma 00} + X_{i}^{\sigma 0\sigma} + X_{i}^{\sigma 0\sigma} + 2 \left( X_{i}^{\sigma 00} + X_{i}^{\sigma 0\sigma} + X_{i}^{\sigma 0\sigma} \right) \right) + (U - 3J_H) \sum_{i,\sigma} \left( X_{i}^{\sigma 00} + X_{i}^{\sigma 0\sigma} + X_{i}^{\sigma 0\sigma} \right),
\]

\[
H_b^{(\lambda)} = \sum_{ij,\sigma} \left( t_{ij} X_{ij}^{000,000} X_{ij}^{000,000} + \bar{t}_{ij} X_{ij}^{000,000} X_{ij}^{000,000} + \bar{t}_{ij} X_{ij}^{000,000} X_{ij}^{000,000} \right),
\]

\[
H_h^{(\lambda)} = \sum_{ij,\sigma} \left( X_{ij}^{000,000} X_{ij}^{000,000} + X_{ij}^{000,000} X_{ij}^{000,000} - X_{ij}^{000,000} X_{ij}^{000,000} - X_{ij}^{000,000} X_{ij}^{000,000} \right),
\]

\[
H_b^{(\gamma)} = \sum_{ij,\sigma} \left( t_{ij} X_{ij}^{000,000} X_{ij}^{000,000} + \bar{t}_{ij} X_{ij}^{000,000} X_{ij}^{000,000} + \bar{t}_{ij} X_{ij}^{000,000} X_{ij}^{000,000} \right),
\]

\[
H_h^{(\gamma)} = \sum_{ij,\sigma} \left( X_{ij}^{000,000} X_{ij}^{000,000} + X_{ij}^{000,000} X_{ij}^{000,000} + X_{ij}^{000,000} X_{ij}^{000,000} \right). 
\]
The technique of Green functions allows us to calculate the energy spectrum within the model, which corresponds to the electronic subsystem of $\Lambda_\gamma C_{00}$ in the case of the electron concentration $n = 1$. One can rewrite the single-particle Green function $\langle a_\alpha | \epsilon^{\dagger}_{\alpha} a_{\alpha} | \rangle$ on the basis of the relation between electronic operators and Hubbard’s $X$-operators:

$$a_{\alpha \gamma} = X_{\alpha \gamma}^{000,100} + X_{\alpha \gamma}^{010,110} + X_{\alpha \gamma}^{001,101} \equiv X_{\alpha \gamma}^{000,100} + Y_{\alpha \gamma},$$

(9)

where the operator $Y_{\alpha \gamma}$ describes the transition processes between doubly occupied Hund’s state and singly occupied state. The processes involving the other types of doubly occupied states, empty states, and states with three or more electrons are improbable due to the energy scaling.

In this way, we obtain the following expression for the single-electron Green function

$$\langle \langle a_{\alpha \gamma} | a_{\alpha \gamma}^{\dagger} \rangle \rangle = \langle \langle X_{\alpha \gamma}^{000,100} | X_{\alpha \gamma}^{000,100} \rangle \rangle +$$

$$+ \langle \langle X_{\alpha \gamma}^{000,100} | Y_{\alpha \gamma}^{+} \rangle \rangle + \langle \langle Y_{\alpha \gamma}^{+} | X_{\alpha \gamma}^{000,100} \rangle \rangle + \langle \langle Y_{\alpha \gamma}^{+} | Y_{\alpha \gamma}^{+} \rangle \rangle.$$

(10)

The equation of motion for the Green function $\langle \langle X_{\alpha \gamma}^{000,100} | X_{\alpha \gamma}^{000,100} \rangle \rangle$ has the form

$$(E + \mu) \langle \langle X_{\alpha \gamma}^{000,100} | X_{\alpha \gamma}^{000,100} \rangle \rangle =$$

$$= \delta_{\mu \lambda} \frac{X_{\alpha \gamma}^{000} + X_{\alpha \gamma}^{000}}{2\pi} + \langle \langle X_{\alpha \gamma}^{000,100} | \sum_{\lambda} H_{\lambda} (\lambda) | X_{\alpha \gamma}^{000,100} \rangle \rangle$$

$$+ \langle \langle X_{\alpha \gamma}^{000,100} | \sum_{\lambda} H_{\lambda} (\lambda) | X_{\alpha \gamma}^{000,100} \rangle \rangle,$$

(11)

and the equation of motion for the Green function $\langle \langle Y_{\alpha \gamma}^{+} | X_{\alpha \gamma}^{000,100} \rangle \rangle$ looks as

$$(E + \mu - U + 3J_H) \langle \langle Y_{\alpha \gamma}^{+} | X_{\alpha \gamma}^{000,100} \rangle \rangle =$$

$$= \langle \langle Y_{\alpha \gamma}^{+} | \sum_{\lambda} H_{\lambda} (\lambda) | X_{\alpha \gamma}^{000,100} \rangle \rangle + \langle \langle Y_{\alpha \gamma}^{+} | \sum_{\lambda} H_{\lambda} (\lambda) | X_{\alpha \gamma}^{000,100} \rangle \rangle.$$

(12)

To obtain the closed system of equations for the Green functions $\langle \langle X_{\alpha \gamma}^{000,100} | X_{\alpha \gamma}^{000,100} \rangle \rangle$ and $\langle \langle Y_{\alpha \gamma}^{+} | X_{\alpha \gamma}^{000,100} \rangle \rangle$, we use the projection procedure similar to that in [28]:

$$\langle \langle X_{\alpha \gamma}^{000,100} | \sum_{\lambda} H_{\lambda} (\lambda) \rangle \rangle = \sum_{\lambda} \epsilon_{\frac{1}{2} \lambda} X_{\alpha \gamma}^{000,100}.$$

(13)

Performing the Fourier transformation, we obtain the Green function in the form:

$$\langle \langle X_{\alpha \gamma}^{000,100} | X_{\alpha \gamma}^{000,100} \rangle \rangle = \frac{X_{\alpha \gamma}^{000} + X_{\alpha \gamma}^{000}}{2\pi} x_{\alpha \gamma}^{000}$$

$$+ \frac{E + \mu - U + 3J_H - \epsilon_{\alpha}^{\pm}(k)}{(E - E_{\lambda}(k)) (E - E_{\gamma}(k))},$$

(14)

where the quasiparticle energy spectrum

$$E_{\alpha \gamma}(k) = -\mu + \frac{U - 3J_H}{2} + \frac{\epsilon_{\alpha}(k) + \epsilon_{\gamma}(k)}{2} +$$

$$\pm \frac{1}{2} \sqrt{(U - 3J_H - \epsilon_{\alpha}^{\pm}(k) + \epsilon_{\gamma}^{\pm}(k))^2 + 4\epsilon_{\alpha}(k)\epsilon_{\gamma}(k)}.$$
\[
\times (X_p^{0\uparrow} + X_p^{0\uparrow} + X_p^{\uparrow\downarrow} + X_{p'}^{\uparrow\downarrow}) + \\
+ (X_{p'}^{0\uparrow\uparrow} + X_{p'}^{1\uparrow\downarrow} + X_{p'}^{\uparrow\downarrow} + X_{p'}^{\downarrow\uparrow}) + \\
- (X_{p'}^{0\uparrow\uparrow} + X_{p'}^{1\uparrow\downarrow} + X_{p'}^{\uparrow\downarrow} + X_{p'}^{\downarrow\uparrow}) - \\
+ (X_{p'}^{0\uparrow\uparrow} + X_{p'}^{1\uparrow\downarrow} + X_{p'}^{\uparrow\downarrow} + X_{p'}^{\downarrow\uparrow}) \\
\]

\[
\tilde{\varepsilon}_k^b = -\frac{i_k}{C_2} \left[ (X_p^{0\uparrow\uparrow} + X_p^{0\uparrow\downarrow} + X_p^{\uparrow\downarrow}) \times \\
\times (X_p^{0\uparrow\uparrow} + X_p^{0\uparrow\downarrow} + X_p^{\uparrow\downarrow}) + \\
+ (X_{p'}^{0\uparrow\uparrow} + X_{p'}^{0\uparrow\downarrow} + X_{p'}^{\uparrow\downarrow}) + \\
- (X_{p'}^{0\uparrow\uparrow} + X_{p'}^{0\uparrow\downarrow} + X_{p'}^{\uparrow\downarrow}) + \\
+ (X_{p'}^{0\uparrow\uparrow} + X_{p'}^{0\uparrow\downarrow} + X_{p'}^{\uparrow\downarrow}) \right], \\
\]

\[
s = \frac{1}{6} (\frac{1}{6} - 12d). \\
\]

Finally in the paramagnetic case at \( n = 1 \), we obtain

\[
\varepsilon^b = \frac{216d^2 - 12d + 1}{24d + 1} t_k + \frac{72d^2}{24d + 1} \tilde{t}_k, \\
\]

\[
\varepsilon^h = \frac{7d^2}{1 - 6d}, \\
\]

\[
\tilde{\varepsilon}^b = \frac{36d^2}{1 - 6d} + \frac{\tilde{t}_k}{2(1 - 6d)}, \\
\]

\[
\tilde{\varepsilon}^h = \frac{24d + 1 - 216d^2}{3(24d + 1)}. \\
\]

In this way, the energy spectrum depends on the concentration of doublons \( d \) (through the dependence of non-operator coefficients). The doublon concentration is determined by the condition

\[
6d = \frac{1}{2N} \sum_k \left( \frac{A_x(k)}{\exp(\frac{E_1(k)}{kT})} + \frac{B_x(k)}{\exp(\frac{E_2(k)}{kT})} + 1 \right), \\
\]

where

\[
A_x(k) = \frac{1}{2} \left( 1 + \frac{U - 3J_H + \varepsilon^h - \varepsilon^b}{\sqrt{(U - 3J_H - \varepsilon^h + \varepsilon^b)^2 + 4\varepsilon^h\varepsilon^b}} \right), \\
B_x(k) = 1 - A_x(k). \\
\]

Using the model rectangular density of states at zero temperature, we obtain

\[
6d = \frac{1}{4\pi} \int_{-\omega}^{\omega} \frac{A_x(\varepsilon) \delta(-E_1(\varepsilon))}{E - E_1(\varepsilon)} d\varepsilon + \\
- \omega \\
\]

\[
\]

\[
\]

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Fig. 2. Energy gap versus the correlated hopping parameter $\tau_1$. The values of $\tau$ are 0; 0.05; 0.1 (starting from the lower curve). The Coulomb interaction parameter $\frac{U-3J_H}{w} = 2$

$$+ \frac{1}{4w} \int_{-w}^{w} \frac{B_{\epsilon}(\epsilon)\Theta(-E_{2}(\epsilon))}{E - E_{2}(\epsilon)} \, d\epsilon,$$

(23)

where $\Theta(-E(\epsilon))$ is the Heaviside theta-function. Solving this equation numerically, we obtain the doublon concentration as a function of the model parameters. To study the metal-insulator transition (MIT) [35–37], we apply the gap criterion

$$\Delta E = E_{2}(-w) - E_{1}(w) = 0.$$  

(24)

At the point of MIT, the concentrations of polar states (holes and doublons) are equal to zero. Thus, for the non-operators coefficients, we have $\varepsilon^b = t_{k}^b$, $\varepsilon^h = 0$, $\tilde{\varepsilon}^b = \frac{t_{k}^b}{2}$, $\tilde{\varepsilon}^h = \frac{t_{k}^b}{3}$. The energy gap satisfies the equation

$$\Delta E = U - 3J_H - \tilde{\omega} - w.$$  

(25)

Here, $w = z|t|(1 - \tau_1)$ and $\tilde{\omega} = z|t|(1 - \tau_1)(1 - 2\tau)$ are the half-bandwidths of the lower and upper subbands, respectively, $z$ is the number of nearest neighbors for a site, $|t|$ is the magnitude of the bare nearest-neighbor hopping integral, $\tau_1$ and $\tau = \frac{t_{ij}'}{|t_{ij}|}$ are the correlated hopping parameters. From relation (25), we obtain that the critical value of intrasite Coulomb interaction parameter equals the sum of the half-bandwidths of quasiparticle subbands.

The analysis of expression (25) allows us to explain the differences of electrical characteristics (of the insulator or metallic state) depending on the correlated hopping strength (see Figs. 2 and 3).

One can see from Fig. 2 that the correlated hopping influences substantially the electrical characteristics of the narrow band material with three-fold orbital degeneracy of the energy levels. Both the filling of the sites involved into the hopping processes (through the first-type correlated hopping) and the neighbor sites (through the second-type correlated hopping) can lead to the appearance of a gap in the energy spectrum and the stabilization of the insulator state. However, the energy gap is opened at a relatively large increase of correlated hopping parameters, which can not be achieved in the compound by a change of external conditions only. Such critical increase of the parameters $\tau_1$ and $\tau$ can be realized at the doping. A distinct picture is observed at a change of the intrasite Coulomb interaction parameter. Under an increase of $(U - 3J_H)/w$ over the critical value (dependent on the correlated hopping strength), the energy gap occurs, and the metal-insulator transition takes place (see Fig. 3). The critical value in the partial case of the model where the quasiparticle subbands have the same widths (in the absence of the correlated hopping) is $(U - 3J_H)/w = 2$, which corresponds to the result of works [28, 38] based on the non-degenerated Hubbard model.

3. Conclusions

Within a version of the triply orbitally degenerate model of the electronic subsystem of a doped fulleride compound considered above, not only the on-site Coulomb correlations but also additional interactions of basic importance, namely the correlated hopping, can be introduced and analyzed. The use of Hubbard representation of $X$-operators appears useful to omit the parts of the
Hilbert space, which are irrelevant at a particular band filling. The ground-state metal-insulator transition in the triply degenerate model of partially-filled doped fulleride band takes place at moderate values of the correlation parameter. In this case, the parameter is a combination of the on-site Coulomb repulsion energy, Hund’s rule coupling, and electron hopping parameters. The correlated hopping of electrons leads to a further localization of current carriers. The influence of the correlated hopping is substantial and makes the estimation of the model parameters from the available spectroscopic data ambiguous. The problem can be resolved by the model parameters from the available spectroscopic data.

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МОТТ-ГАББАРДІВСЬКА ЛОКАЛІЗАЦІЯ В МОДЕЛІ ЕЛЕКТРОННОЇ ПІДСИСТЕМИ ЛЕГОВАНОГО ФУЛЕРИДУ

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Резюме

Досліджено конфігураційне представлення мікроскопічної моделі електронної підсистеми легованого фуллеренду з урахуванням трикритального орбітального виділення електронних статей. З використанням методу функцій Гітана розраховано енергетичний спектр моделі при заповненні зони $n = 1$, яку відповідає сполукам $\text{AC}_60$. Обґрунтовано можливі кореляційні індукуваний перехід діелектрик–метал у рамках даної моделі.