Self-consistent approach for thermodynamics of a simplified pseudospin-electron model

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We present the method of the self-consistent calculation of thermodynamical and correlation functions. This approach is based on the GRPA (generalized random phase approximation) scheme with the inclusion of the mean field corrections. Investigation of a pseudospin-electron model (PEM) within the framework of the presented method shows that interaction between the electron and pseudospin subsystems leads to the possibility of either first or second order phase transitions between different uniform phases (bistability) as well as between the uniform and the chess-board ones. In the regime \( n = \text{const} \), an instability with respect to phase separation in the electron and pseudospin subsystems can take place.

Key words: pseudospin-electron model, chess-board phase, phase transitions, local anharmonicity, high-Tc superconductors

PACS numbers: 71.10.Fd, 71.38.+i, 77.80.Bh, 63.20.Ry

1 Introduction

The crystals with the high-temperature superconductivity (HTSC) investigated intensively during the last ten years possess a wide spectrum of the interesting physical properties. The variety of effects which are realized only separately in the case of other types of crystals is characteristic of these systems. The strong electron correlation, related to the interaction of the Hubbard type in the conducting bands formed mainly by the superconducting Cu\(_2\)-O\(_2\) plains, can be pointed out as one of the reasons of this unique situation.

Another significant feature connected with the dynamics of HTSC crystals is the presence of strongly anharmonic elements of the structure. As it is known, it can be a source of instabilities of various type.

Among the most frequently investigated HTSC crystals are the crystals of the YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) group. The unit cell of these compounds contains, besides two superconducting planes, the chain (at the \( \delta \ll 1 \) composition) elements Cu\(_1\)-O\(_1\), connected by Cu\(_1\)-O\(_4\)-Cu\(_2\) bridges with conducting plains through the apical oxygen ions O\(_4\). The vibrations of these ions along the c-axis (perpendicularly to the plains) exhibit a strong anharmonicity. Much evidence exists in support of this concept. One can mention the results of experimental investigations (EXAFS data [1,2], Raman scattering [3-6], dielectric measurements [7,8]) where the two equilibrium positions of O\(_4\) ion (two different values of the \( R_{O_4-Cu_2} \) distance) have been observed that can point out to the presence of the local double-minimum potential well. Similar conclusions were made based on consideration the local polaron phenomena [9,10], electron transfer processes through O\(_4\) ions [11,12] or bistabilities in the normal phase temperature region [13].

Besides, a connection between positions of O\(_4\) ions and the energy of electron states in Cu\(_2\)-O\(_2\) plains plays an important role in YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) crystals. The data given in [14] point to the existence of a significant correlation between the occupancy of electron states of the Cu\(_2\) ion and the \( R_{O_4-Cu_2} \) distance as well as to the decrease of this distance at the transition from the metallic orthorombic phase to the semiconducting phase (that takes place at \( \delta > \delta^* = 0.55 \)). These and other similar facts suggest the presence of a large electron-vibrational coupling.

By now, the description of the locally anharmonic subsystem in HTSC crystals develops with the use of two different approaches. The first one, which is chronologically older, is based on the phonon anharmonic model \( \varphi^3 \) [15], or, more recently, on the model \( \varphi^3 + \varphi^4 \) [13]. This approach was used while consideration the polaron effect [16] and also while investigating the effect of anharmonicity on the superconducting transition temperature and on the possibility of the modulated (of the CDW type) phase creation [17].

In the second approach, which is more appropriate at the strong electron-vibrational coupling and in the cases when two equilibrium positions of anharmonic ions really exist, the pseudospin formalism is used; the pseudospin variable \( S_i^z = \pm 1/2 \) defines these two positions. This scheme being applied to the YBa\(_2\)Cu\(_3\)O\(_{7-\delta}\) type systems started from works [18] and [19].

On the basis of the second model which was called as pseudospin-electron model (PEM) a possible connection between the superconductivity and lattice instability of the ferroelectric type in HTSC has been discussed [19,20]. The description of the electron spectrum and the electron statistics of the PEM was given in [21] in the framework of the temperature two-time Green function method in the Hubbard-I approximation.
A series of works has been carried out in which the pseudospin \( (S^2 S^z) \), mixed \( \langle S^z n \rangle \) and charge \( \langle nn \rangle \) correlation functions were calculated. It has been shown with the use of the generalized random phase approximation (GRPA) [22] in the limit of infinite single-site electron correlations \( (U \to \infty) \) [23,24], that there exists a possibility of divergences of these functions at some values of temperature. This effect was interpreted as a manifestation of dielectric instability or ferroelectric type anomaly. The tendency to the spatially modulated charge and pseudospin ordering at the certain model parameter values was found out.

On the other hand, the case of absence of the term describing electron transfer in Cu2O2 plains \( (t_{ij} = 0) \) with the inclusion of the direct interaction between pseudospins was considered within the mean field approximation [25,26] (see also the short review in [27]). The first or second order phase transitions with the jumps of \( \langle S^z \rangle \) and electron concentration \( n \) values in the \( \mu = \text{const} \) regime were obtained. An instability with respect to phase separation in the electron and pseudospin subsystems can take place in regime \( n = \text{const} \).

The analysis of ferroelectric type instabilities in the two-sublattice model of the apex oxygen subsystem in high temperature superconducting systems has been made [28]. The influence of oxygen nonstoichiometry on localization of apex oxygen in YBa2Cu3O7-\( \delta \) type crystals was studied in the work [29].

In the present work we propose the self-consistent scheme for calculation of mean values of pseudospin and electron number operators, grand canonical potential as well as correlation functions for the case of the \( U = 0 \) limit (the simplified PEM). The approach is based on the GRPA with the inclusion of the mean field type contributions coming from effective pseudospin interactions via conducting electrons [30]. The main attention is paid to the thermodynamics of phase transitions. The possibilities of phase separation and chess-board phase appearance are investigated.

## 2 Pseudospin-electron model

Hamiltonian of the pseudospin-electron model has the following form:

\[
H = H_0 + \sum_{ij\sigma} t_{ij} b_{ij\sigma}^+ b_{j\sigma},
\]

\[
H_0 = \sum_i \left\{ U n_{i\uparrow} n_{i\downarrow} - \mu \sum_{\sigma} n_{i\sigma} + g \sum_{\sigma} n_{i\sigma} S^z_i - h S^z_i \right\},
\]

where the strong single-site electron correlation \( U \), interaction with the anharmonic mode \( (g\text{-term}) \) and the energy of the anharmonic potential asymmetry \( (h\text{-term}) \) are included in the single-site part; \( \mu \) is the chemical potential. The second term in the Hamiltonian describes the electron hopping from site to site (the electron transfer parameter \( t_{ij} \)).

The formalism of electron annihilation (creation) operators \( a_{i\sigma} = b_{i\sigma} P_i^+ \), \( \tilde{a}_{i\sigma} = b_{i\sigma} P_i^- \) \( (P_i^+ = \frac{1}{2} \pm S^z_i) \) acting at a site with the certain pseudospin orientation is introduced. The calculation is performed in the strong coupling case \( (g \gg t) \) using of single-site states as the basic one.

\[
H_0 = \sum_{i} \{ \varepsilon (n_{i\uparrow} + n_{i\downarrow}) + \varepsilon (\tilde{n}_{i\uparrow} + \tilde{n}_{i\downarrow}) - h S^z_i \},
\]

\[
H_{\text{int}} = \sum_{ij\sigma} t_{ij} (a_{i\sigma}^+ a_{j\sigma} + a_{i\sigma}^+ \tilde{a}_{j\sigma} + a_{i\sigma} \tilde{a}_{j\sigma} + a_{i\sigma}^+ \tilde{a}_{j\sigma}^+).
\]

Here \( \varepsilon = -\mu + g/2, \ \tilde{\varepsilon} = -\mu - g/2 \) are energies of the single-site states. We consider here the simplified PEM (the case \( U = 0 \)).

Expansion of the calculated quantities in terms of electron transfer leads to the infinite series of terms containing the averages of the \( T \)-products of the \( a_{i\sigma}, \tilde{a}_{i\sigma} \) operators. The evaluation of such averages is made using the corresponding Wick’s theorem. The results are expressed in terms of the products of nonperturbed Green functions and averages of a certain number of the projection operators \( P_i^\pm \) which are calculated by means of the semi-invariant expansion [30].

Single-electron Green function (calculated in Hubbard-I type approximation) may be written as the following chain diagram:

\[
\langle S^z_i \rangle = \sum_{\omega_n} \langle g_i(\omega_n) \rangle = \sum_{\omega_n} \frac{P_i^+}{i \omega_n - \varepsilon} + \frac{P_i^-}{i \omega_n - \tilde{\varepsilon}},
\]

where we use the following diagrammatic notations:

\[
\begin{align*}
\bigcirc = & S^z_i, & -\longrightarrow = g_i(\omega_n), & \bigcirc \bigcirc \bigcirc = \frac{P_i^+}{i \omega_n - \varepsilon} + \frac{P_i^-}{i \omega_n - \tilde{\varepsilon}}, & \text{nonperturbed electron Green function}
\end{align*}
\]

Here \( \omega_n \) is the Matsubara frequency.

The wavy line is the intersite hopping \( t_{ij} \). Semi-invariants are represented by ovals and contain the \( \delta \)-symbols on site indexes. In the spirit of the traditional mean field approach [30] the renormalization of the basic semi-invariant by the insertion of independent loop fragments is taken into account in (1).
The diagrammatic series for the electron concentration mean value is the following:

\[ \langle n_i \rangle = \square - \frac{1}{2!} \bigoplus + \ldots \]  

where \( \square = n_i \), \( \bigoplus = \frac{1}{\omega_n - \varepsilon^\alpha} \), \( P_0^\alpha = (P_i^+, P_i^-) \).

Hence, with respect to the GRPA scheme an influence of the internal effective self-consistent field on pseudospins is taken into account by means of the inclusion of the mean field type contributions into the expressions for all thermodynamic quantities. In the approach presented the correlation functions are calculated consistently with thermodynamical functions.

The consistency of the expressions \( (3) \)–\( (6) \) can be checked explicitly [30] using the thermodynamical relations:

\[ \frac{d\Omega}{d(-\mu)} = \langle n \rangle, \quad \frac{d\Omega}{d(-h)} = \langle S^z \rangle, \quad \frac{d\langle S^z \rangle}{d(\beta h)} = \langle S^z S^z \rangle_{q=0}. \]

At high temperatures eq. \( (3) \) possesses only the uniform solution \( \langle S^z \rangle = \langle \hat{S}^z \rangle \). The phase transitions between uniform phases with different pseudospin mean values \( \langle S^z \rangle \) were analyzed in [30]. For the first time the possibility of the dielectric instabilities has been done for the PEM in the limit of the strong electron correlation \( (U \to \infty) \) in [23,24]. Results of ref. [30], where the opposite case of \( U \to 0 \) was analyzed, are in good agreement with the exact ones for the \( U \to 0 \) PEM in the limit of infinite spatial dimension [31]. At the same time, a complete description of such transitions was obtained in [25,26] for the PEM with direct interaction between pseudospins \( (t_{ij} = 0 \) limit).

On the other hand, the solution of eq. \( (6) \) for pseudospin correlator has the form

\[ \langle S_i^z S_j^z \rangle_q = \frac{1}{4(1 - \langle S^z \rangle^2)} \left( 1 + \prod_q \left( \frac{1}{4} - \langle S^z \rangle^2 \right) \right), \]

where \( \prod_q \) characterize an interaction between pseudospins via electron subsystem:

\[ \prod_q = \sum_{\alpha, \beta} (-1)^{\alpha + \beta} \bigoplus, \]

and its singularities

\[ 1 + \prod_q \left( \frac{1}{4} - \langle S^z \rangle^2 \right) = 0 \]

gives the points of the instability of uniform phase with respect to fluctuations with wave vector \( q \). The typical dependence of the temperature of instability on the fluctuation wave vector \( q \) for square lattice is shown in figure 1 and one can see that for some model parameter values uniform phase become unstable with respect to fluctuations with \( q = (\pi, \pi) \) (chess-board phase).
So, below we shall take into account the possibility of the chess-board phase appearance in our analysis.

3 The chess-board phase

To take into account the modulation of the pseudospin and electron distribution we introduce two kinds of sites: the \( \langle S^1_i \rangle \) corresponds to the one sublattice, and the \( \langle S^2_i \rangle \) to the other one. The nearest–neighbour hopping exists only between the sublattices.

Single–electron Green functions \( \langle \rangle \) in this case are equal:

\[
\begin{align*}
G_{1k}(\omega_n) &= \frac{g_1(\omega_n)}{1 - t^2_k g_1(\omega_n) g_2(\omega_n)}, \\
G_{2k}(\omega_n) &= \frac{g_2(\omega_n)}{1 - t^2_k g_1(\omega_n) g_2(\omega_n)},
\end{align*}
\]

where \( g_1(\omega_n) \) and \( g_2(\omega_n) \) are nonperturbed Green functions for the sublattices 1 and 2 respectively. The poles of functions \( G_k(\omega_n) \) determine the single–electron spectrum. The equation for the spectrum has the form:

\[
x^4 - (g^2/2 + t_k^2)x^2 - gt_k^2(\langle S^1_i \rangle + \langle S^2_i \rangle)x + g^4/16 - g^2t^2_k(\langle S^1_i \rangle \langle S^2_i \rangle) = 0,
\]

where \( x = i\omega_n + \mu \).

The roots \( \varepsilon_1(t_k) \geq \varepsilon_2(t_k) \geq \varepsilon_3(t_k) \geq \varepsilon_4(t_k) \) of the equation \( \langle \rangle \) form four subbands. The widths of subbands depend on the mean values of pseudospins.

The branches \( \varepsilon_1(t_k), \varepsilon_2(t_k) \) on the one side and \( \varepsilon_3(t_k), \varepsilon_4(t_k) \) on the other one coincide at \( t_k = 0 \) \( \varepsilon_{1.2}(t_k = 0) = g/2, \varepsilon_{3.4}(t_k = 0) = -g/2 \) and form two pairs of bands which are always separated by gap.

The equations for pseudospin mean values \( \langle \rangle \) can be written now in the form:

\[
\langle S^1_i \rangle = \frac{1}{2} \tanh \left\{ \frac{\beta}{2} (h + \alpha^1_2 - \alpha^1_1) + \ln \frac{1 + e^{-\beta \varepsilon}}{1 + e^{-\beta \bar{\varepsilon}}} \right\},
\]

\( l = 1, 2; \) where expressions for the effective self-consistent fields are

\[
\begin{align*}
\alpha^1_2 - \alpha^1_1 &= \frac{2}{N} \sum_k t^2_k (\varepsilon - \bar{\varepsilon}) \sum_{i=1}^4 A^1_i n[\varepsilon_i(t_k) - \mu], \\
A^1_i &= \langle \varepsilon_i(t_k) + g \langle S^1_i \rangle \rangle / (\langle \varepsilon_i(t_k) - \varepsilon_j(t_k) \rangle (\langle \varepsilon_i(t_k) - \varepsilon_p(t_k) \rangle (\langle \varepsilon_i(t_k) - \varepsilon_m(t_k) \rangle)),
\end{align*}
\]

\( i \neq j, p, m; \quad l \neq l'. \)

Expression for electron number mean value follows from \( \langle \rangle \):

\[
\langle n_1 + n_2 \rangle = \frac{2}{N} \sum_k \sum_{i=1}^4 n[\varepsilon_i(t_k) - \mu] - 2 \left[ (\langle P^+_1 \rangle + \langle P^+_2 \rangle) n(\varepsilon) + (\langle P^-_1 \rangle + \langle P^-_2 \rangle) n(\varepsilon) \right],
\]

and grand canonical potential \( \langle \rangle \) can be written in the analytical form, also:

\[
\Delta \Omega = -\frac{2}{N\beta} \sum_k \ln \frac{\prod_{i=1}^4 \cosh \left[ \frac{\beta}{2} (\varepsilon_i(t_k) - \mu) \right]}{(\cosh \frac{\beta}{2} \varepsilon)^2 (\cosh \frac{\beta}{2} \bar{\varepsilon})^2} + \sum_{l=1,2} \langle S^1_i \rangle (\alpha^1_2 - \alpha^1_1) + \sum_{l=1,2} \left[ -\frac{1}{\beta} \ln \cosh \left\{ \frac{\beta}{2} (h + \alpha^1_2 - \alpha^1_1) + \ln \frac{1 + e^{-\beta \varepsilon}}{1 + e^{-\beta \bar{\varepsilon}}} \right\} \right] + \frac{1}{\beta} \ln \cosh \left\{ \frac{\beta}{2} h + \ln \frac{1 + e^{-\beta \varepsilon}}{1 + e^{-\beta \bar{\varepsilon}}} \right\}.
\]

4 Numerical results

In the investigation of equilibrium conditions we shall separate two different regimes in which the system can exist:

a) The \( \mu = \text{const} \) regime; it is supposed that the electron states of other structure elements, which are not included explicitly into the PEM, play a role of a thermostat, that ensures a constant value of the chemical potential \( \mu \) (despite the possible changes of temperature, field \( h \) and other characteristics of the model). In this case the minimum of the grand canonical potential \( \Omega \) is a condition of thermodynamical equilibrium;
b) the regime \( n = \text{const} \); this situation is more customary at the consideration of electron systems and it means that the chemical potential is now the function of \( T, h \) etc. and depends on the electron concentration. The minimum of the free energy \( F = \Omega + \mu N \) is the equilibrium condition in this case.

4.1 \( \mu = \text{const} \) regime

For this regime the equilibrium is defined by the minimum condition of \( \Omega \) (15) that form the equations for pseudospin mean values (12) and expression for electron concentration (14). The calculated field dependences of \( \langle S_z^1 - S_z^2 \rangle \) (order parameter for chess-board phase) and grand canonical potential (which are determined by the solutions of the eqs. (12)) are presented in figure 2 for \( g \gg W \) and low temperature.

![Figure 2](image1.png)

Figure 2. Field dependence of the grand canonical potential and order parameter \( \langle S_z^1 - S_z^2 \rangle \) \( (T = 0.005, \mu = -0.36, W = 0.2, g = 1) \). Dotted and solid lines correspond to the uniform and chess-board phases, respectively.

Comparison of the grand canonical potential \( \Omega \) values for uniform and chess-board phases leads to the conclusion that the modulated phase is thermodynamically stable at intermediate values of \( h \) parameter in the region between points a and b. These points correspond to the first and second order phase transition, respectively. The rapid jump-like change of order parameters is accompanied by the rapid changes of the subbands widths and, as a result, electron concentration.

The resulting phase diagram \( \mu - h \) at the low temperature is shown in figure 3.

![Figure 3](image2.png)

Figure 3. \( \mu - h \) phase diagram \( (T = 0.005, W = 0.2, g = 1) \). I - uniform phase, II - chess-board phase. Dashed lines - first order phase transitions between the uniform phases with different pseudospin mean values. Dotted lines - first order phase transitions between the uniform and chess-board phases. Solid lines - second order phase transitions.

The transitions between uniform phases with different pseudospin mean values as well as corresponding electron concentrations (bistability), which is of the first order, takes place when the chemical potential \( \mu \) is placed within the \( \varepsilon_1, \varepsilon_2 \) and partially \( \varepsilon_3, \varepsilon_4 \) bands. The transitions between the uniform and modulated phases are of the first or second order and can be realized in the case when \( \mu \) is placed in \( \varepsilon_2 \) and \( \varepsilon_3 \) bands or between them. The chess-board phase exists as intermediate one between the uniform phases with different \( \langle S_z^2 \rangle \) and \( n \) values.

The \( T - h \) phase diagram is shown in figure 4.

![Figure 4](image3.png)

Figure 4. Phase diagram \( T - h \) \( (\mu = -0.36, W = 0.2, g = 1) \). I - uniform phase, II - chess-board phase. Dashed lines - first
order phase transitions between the different uniform phases (bistability). Dotted line – first order phase transitions between the uniform and chess-board phases. Solid lines – second order phase transitions.

With the temperature increase the first order phase transition between the uniform and chess-board phases transforms into the first order phase transition between uniform phases and, finally, disappear in the critical point \( \theta_c \).

The diagram \( T - h \) shows the possibility of the first order phase transitions between uniform phases and either first or second order ones between the uniform and chess-board phases at the change of temperature.

In figures 2 and 4 the case when the chemical potential is placed in the lower band is presented. If the chemical potential is placed in the upper band our results transform according to the symmetry of the Hamiltonian: \( \mu \rightarrow -\mu, h \rightarrow 2g - h, n \rightarrow 2 - n, S^z \rightarrow -S^z \).

4.2 \( n = \text{const} \) regime

In the regime of a fixed value of electron concentration the equilibrium is defined by the minimum of free energy \( F = \Omega + \mu N \). This condition form a set of equations (12) and (14) for the pseudospin mean values and chemical potential. The obtained dependences of \( F \) and \( \mu \) on the electron concentration are presented in figure 5.

One can see the regions with \( d\mu/dn \leq 0 \) where states with a homogeneous distribution of particles are unstable, which corresponds to the phase separation into the regions with different phases (the uniform and chess-board ones in this case) and with different electron concentrations and pseudospin mean values (that is, different occupancies of particle positions in the anharmonic potential wells). In the phase separated region the free energy, as a function of \( n \), deflects up and electron concentrations in the separated phases are determined by the tangent line touch points.

On the base of the obtained results the phase diagram \( n - h \) was constructed (figure 6).

5 Conclusions

On the basis of the presented above self-consistent scheme for calculation of the correlation and thermodynamical functions, the energy spectrum, thermodynamics of phase transitions, possibility of phase separations as well as appearance of the chess-board phase have been investigated. The corresponding phase diagrams were build. Such comprehensive analysis of the thermodynamics of the considered simplified PEM became possible due to the generalization of the traditional GRPA approach (in which, on the basis of calculation of the \( \langle S^z S^z \rangle \) correlation function, only the assumption about the chess-board phase appearance may be done).
The obtained phase diagrams remind the situation know for the Falicov-Kimball (FK) model (this model is close to PEM but differ in the thermodynamic equilibrium conditions) with a rich phase diagram. However, contrary to this model, an existence of the phase transitions between uniform phases is possible in our case. This results from the another regime of thermodynamic averaging (fixation of $h$ field which is an analogous to the chemical potential for ions in the FK model).

The study of the thermodynamics of the PEM within the framework of the presented above approach provides reason enough to conclude that on the basis of this model one can describe the phase transitions and instabilities in the HTSC of the $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ type. Among them we should list the bistability effects (at the change of temperature), separation into phases with different electron concentrations, pseudospin orientations (that corresponds to different localization of particles in the anharmonic potential wells). In general, it is in agreement with the picture observed experimentally (some publications in this field were quoted above).

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