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STRUCTURAL PHASE TRANSITIONS IN THE TWO-SUBLATTICE PSEUDOSPIN-ELECTRON MODEL OF HIGH TEMPERATURE SUPERCONDUCTING SYSTEMS

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Структурні фазові переходи в двопідгратковій псевдоспін-електронній моделі високотемпературних надпровідників

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Анотація. Діелектричні властивості двопідграткової моделі Хаббарда з локальним ангармонізмом та прямою взаємодією між псевдоспінами розглянуті в наближенні середнього поля та однопетлевому наближенні. Така модель може бути використана для опису діелектричних властивостей взовж осі c надпровідників типу YBaCuO (псевдоспіни описують ангармонічні коливання вершинних киснів O4). Поведінку поперечної діелектричної сприйнятливісті $\chi_\perp$ досліджено в наближенні невзаємодіючих кластерів. Визначено значення параметрів моделі, при яких мають місце фазові переходи в стан з некомпенсованим впорядкуванням псевдоспінів. Запропоновано метод усунення додаткових (нефізичних) фазових переходів в однопетлевому наближенні.

Structural phase transitions in the two-sublattice pseudospin-electron model of high temperature superconducting systems

I.V.Stasyuk, O.D.Danyliv

Abstract. The dielectric properties of two-sublattice Hubbard model with local anharmonicity and direct interaction between pseudospins are considered within mean field approximation and one-loop approximation. Such a model can be used for the description of dielectric properties of YBaCuO-type superconductors along c-axis (the pseudospins represent an anharmonic motions of apical oxygen O4). The behaviour of the the transverse dielectric susceptibility $\chi_\perp$ is considered in the non-interacting clusters approximation. The regions of model parameters, where phase transitions into phases with noncompensated ordering of pseudospins are established. It is proposed the method how to avoid the additional (nonphysical) phase transition within one-loop approximation (the first order of high-density expansion method).

1. Introduction

The Hubbard model was one of the models to be proposed for the description the behaviour of high temperature superconductors (HTSC). It describes the electrons (holes) transfer in CuO planes of copper-oxygen HTSC taking into account the strong correlation of electrons (holes) on the sites in the superconducting planes. It is simplification of Emery [1] model which more fully describes electron properties of CuO layers. The analysis of EXAFS spectra and Raman spectra for the most studied superconductor YBaCuO [2–4] manifests in the existence of a strong anharmonic double potential well of apex oxygen O4. This conclusion agrees with experiments on the diffraction of X-rays [5]. The role of apex oxygen vibrations (which usually described by the pseudospin formalism) in the superconducting phase transitions can be decisive [6]. In the last years the pseudospin-electron model was proposed in which the Hubbard correlation is supplemented by electron-pseudospin interaction on the site. The dielectric properties and electron spectrum of this pseudospin-electron model were studied within the Hubbard-I approximation [7] and GRPA (generalized random phase approximation) [9]. It was shown that the interaction of electrons with anharmonic vibrational modes leads to the appearance of extra Hubbard sub-bands and causes their additional narrowing. The investigation performed within the mean field approximation [8] gave evidence for the possibility of the phase transition into a charge-ordered state at electron concentration $n \sim 1$. It was pointed that an interaction between electron subsystem and pseudospin one leads to the appearance of an effective Vn_in_j type term. It is responsible for a phase transition with a modulation of electron concentration. The results obtained in GRPA confirm it. Moreover, it was shown that there is a phase transition in the centre of the Brillouin zone at low electron concentrations. This transition manifests itself in the divergency of dielectric susceptibility $\chi_\perp$ at certain value of temperature.

In some experiments YBaCuO was found to be both pyroelectric and piezoelectric, implying the existence of macroscopic polarization directed along the c-axis [10]. In order to explain such type behaviour in the framework of pseudospin-electron model it is important to take into account the interaction between pseudospins. This interaction can lead to ordering of pseudospins and appearance of a ferroelectric phase [11].

In this paper we investigate the dielectric and thermodynamical properties of the two-sublattice pseudospin-electron model. We shall restrict our consideration to the case of zero electron hopping. In Sec.2 we introduce the Hamiltonian of the model. In Sec.3 we calculate the transverse

dielectric susceptibility in the noninteracting clusters approximation. The thermodynamical properties and susceptibility in the presence of pseudospin-pseudospin interaction are considered within the mean field approximation (MFA) in Sec.4. The improving of MFA results (one-loop approximation) is given in Sec.5. Finally, in Sec.6, we present a summary of results and conclusions.

2. The Hamiltonian of two-sublattice model.

The Hamiltonian of the model with lattice local anharmonicity written with the use of pseudospin formalism has the form

$$\begin{aligned} H &= H_e + H_s + H_{e-s} + H_{s-s}, \qquad (1)\\ H_e &= -\mu \sum_{i,s} (\hat{n}^s_{i1} + \hat{n}^s_{i2}) + U \sum_i (\hat{n}^\uparrow_{i1}\hat{n}^\downarrow_{i1} + \hat{n}^\uparrow_{i2}\hat{n}^\downarrow_{i2}),\\ H_s &= -h \sum_i (S^z_{i1} - S^z_{i2}) - \Omega \sum_i (S^x_{i1} + S^x_{i2}),\\ H_{e-s} &= g \sum_{i,s} (\hat{n}^s_{i1} S^z_{i1} - \hat{n}^s_{i2} S^z_{i2}),\\ H_{s-s} &= -J \sum_i S^z_{i1} S^z_{i2} - \frac{1}{2} \sum_{i,i'} \sum_{\alpha,\beta} J^{\alpha\beta}_{ii'} S^z_{i\alpha} S^z_{i'\beta}. \end{aligned}$$

Here, H_e is the Hubbard Hamiltonian without term describing transfer of electrons, H_s is the pseudospin part of the Hamiltonian, H_{e-s} and H_{s-s} are terms describing the electron-pseudospin and the pseudospin-pseudospin interactions respectively. $\hat{n}^s_{i\alpha}$ is the operator of occupation number of electrons with spin s and $S^z_{i\alpha}$ stands for the operator of the pseudospin at the i cell in α plane ($\alpha = 1, 2$ in the two-sublattice case). h describes the asymmetry of site potential (this parameter has opposite signs in sublattices 1 and 2), Ω is the tunnelling splitting of the vibrational mode. The interaction $-JS^z_{i1}S^z_{i2}$ inside one cell clusters is separated. We consider two different variants of model realization: $\mu = const, n = n(\mu)$ and $n = const, \mu = \mu(n)$ with the purpose of elucidating the role of structural elements or electron bands which are not explicitly included into model but can hold the fixed value of the chemical potential. Hamiltonian (1) is invariant with respect to the transformation $\hat{n}^s_{i\alpha} \to 1 - \hat{n}^s_{i\alpha}$, $h \to 2g - h$, $\mu \to -\mu - U$. It allows us to use (1) for the description of hole-pseudospin system as well.

Let us introduce the cluster basis of states $|R_i\rangle \equiv |n^\uparrow_{i1}, n^\downarrow_{i1}, n^\uparrow_{i2}, n^\downarrow_{i2}\rangle \oplus |S^z_{i1}, S^z_{i1}\rangle \equiv |n^\uparrow_{i1}, n^\downarrow_{i1}, n^\uparrow_{i2}, n^\downarrow_{i2}, S^z_{i1}, S^z_{i1}\rangle$ (here $n^\uparrow_{i\alpha}, n^\downarrow_{i\alpha}$ denote the eigenval-

ues of operators $\hat{n}^{\uparrow}_{i\alpha}$ and $\hat{n}^{\downarrow}_{i\alpha}$) which consists of sixty-four state vectors

$$\begin{aligned}
|1\rangle &= |0,0,0,0,\uparrow,\uparrow\rangle \\
|2\rangle &= |0,0,0,0,\uparrow,\downarrow\rangle \\
|3\rangle &= |0,0,0,0,\downarrow,\uparrow\rangle \\
|4\rangle &= |0,0,0,0,\downarrow,\downarrow\rangle \\
|5\rangle &= |0,0,0,1,\uparrow,\uparrow\rangle \\
&\dots \\
|64\rangle &= |1,1,1,1,\downarrow,\downarrow\rangle.
\end{aligned} \tag{2}$$

On this basis the Hamiltonian (1) without interaction $J^{\alpha\beta}_{ii'}$ between clusters has a block diagonal form with size of blocks 4×4. Only the pseudospin part of Hamiltonian is non-diagonal. The $i-th$ block, which corresponds to a fixed electron configuration $|K_i\rangle \equiv |n^{\uparrow}_{i1}, n^{\downarrow}_{i1}, n^{\uparrow}_{i2}, n^{\downarrow}_{i2}\rangle$, has the following form

$$\begin{pmatrix}
\varepsilon^{K_i}_{el} + \frac{g}{2}(n_{i1}-n_{i2}) - \frac{J}{4} & -\frac{\Omega}{2} & -\frac{\Omega}{2} & 0 \\
-\frac{\Omega}{2} & \varepsilon^{K_i}_{el} + \frac{g}{2}(n_{i1}+n_{i2}) + \frac{J}{4} - h & 0 & -\frac{\Omega}{2} \\
-\frac{\Omega}{2} & 0 & \varepsilon^{K_i}_{el} - \frac{g}{2}(n_{i1}+n_{i2}) + \frac{J}{4} + h & -\frac{\Omega}{2} \\
0 & -\frac{\Omega}{2} & -\frac{\Omega}{2} & \varepsilon^{K_i}_{el} - \frac{g}{2}(n_{i1}-n_{i2}) - \frac{J}{4}
\end{pmatrix}
\begin{matrix}
|K_i,\uparrow,\uparrow\rangle \\ |K_i,\uparrow,\downarrow\rangle \\ |K_i,\downarrow,\uparrow\rangle \\ |K_i,\downarrow,\downarrow\rangle.
\end{matrix} \tag{3}$$

Here $n_{i\alpha}$ is the eigenvalue of operator $\hat{n}^{\uparrow}_{i\alpha} + \hat{n}^{\downarrow}_{i\alpha}$, and $\varepsilon^{K_i}_{el}$ is the electron contribution to the energy of the $|K_i\rangle$ state.

In the case $J = 0$ and $J^{\alpha\beta}_{ii'}$, the general problem can be reduced to the problem of two noninteracting sublattices. This corresponds to the case considered in [7].

3. Dielectric susceptibility along c-axis in the absence of the pseudospin-pseudospin interaction.

Let us consider the simplified case $J^{\alpha\beta}_{ii'} = 0$. Then Hamiltonian (1) describes the behaviour of the system of "noninteracting" clusters. The coupling between them exists only through the common reservoir of charge carriers (described by chemical potential μ). That is why the case $J = 0$ is not reduced exactly to the one-sublattice case [9]. We included in the Hamiltonian a part which describes the influence of an electric field perpendicular to the planes:

$$H_{int} = -E_{\perp} \sum_i P^z_i, \tag{4}$$

where P^z_n is the operator of polarization along the direction of the field. It is given by the formula

$$P^z_i = d_s(S^z_{i1} + S^z_{i2}) + d_e(n_{i1} - n_{i2}), \tag{5}$$

where d_s is the dipole moment connected with pseudospin flipping (transition of oxygen atom from one minimum of double well potential to another); d_e is the dipole moment of electron charge transfer between planes. Then the transverse dielectric susceptibility is given by

$$\chi_{\perp} = \frac{1}{v_c N} \frac{\partial \langle \sum_i P^z_i \rangle_{H+H_{int}}}{\partial E_{\perp}}_{|E_{\perp}=0} = \frac{1}{v_c} \frac{\partial \langle\, P^z \rangle_{H+H_{int}}}{\partial E_{\perp}}_{|E_{\perp}=0}, \tag{6}$$

here N is the total number of sites, v_c is the volume of a primitive cell.

In order to understand the possible temperature behaviour of $\chi_{\perp}$, firstly, we consider the ground state of the system at the different values of model parameters. Fig.1 illustrates the ground state diagram for $\Omega = 0$, $J > 0$, $U \to \infty$. The solid lines on this figure separate regions with different ground states. The chemical potential changes along the AC_1DE segment at $n \sim 0$ and along ABCE at $n \sim 2$. One can draw a line parallel to the h-axis to consider the $\mu = const$ case. The calculations show that the eigenvalues of the polarization operator in the ground state have a nonzero values at $J > 0$ only inside the region KHFG; its width is proportional to J (at $J < 0$ this region is absent). Inside region BC_1KH the ground state is doubly degenerate and is described by the state vectors $|R_i\rangle = |0,0,0,0,\uparrow,\uparrow\rangle$, $|0,0,0,0,\downarrow,\downarrow\rangle$. In FGDC region the states with the lowest energy are $|K_i,\uparrow,\uparrow\rangle$ and $|K_i,\downarrow,\downarrow\rangle$; in region $BCDC_1$ the ground state is four-fold degenerate. The electron part of the ground state dipole moment is different from zero only inside the region $BCDC_1$. The susceptibility $\chi_{\perp}$ is proportional to $e^{-\beta\alpha}$ ($\alpha > 0$) outside of those regions and it is obeyed the $\frac{1}{T}$ law inside of them:

$$n = const$$

$$\begin{aligned}
&\boxed{\text{I}}, \boxed{\text{IV}} && \chi_{\perp} = \frac{\beta}{v_c} \frac{(2-n)n}{2} d_e^2; \\
&\boxed{\text{II}} && \chi_{\perp}(0 < n < 1) = \frac{\beta}{v_c} n (d_s - d_e)^2, \\
& && \chi_{\perp}(1 < n < 2) = \frac{\beta}{v_c} (2-n)(d_s - d_e)^2; \\
&\boxed{\text{III}} && \chi_{\perp}(0 < n < 1) = \frac{\beta}{v_c} \{ n(d_s - d_e)^2 + (1-n) d_s^2 \}, \\
& && \chi_{\perp}(1 < n < 2) = \frac{\beta}{v_c} \{ (2-n)(d_s - d_e)^2 + (n-1) d_s^2 \}.
\end{aligned} \tag{7}$$

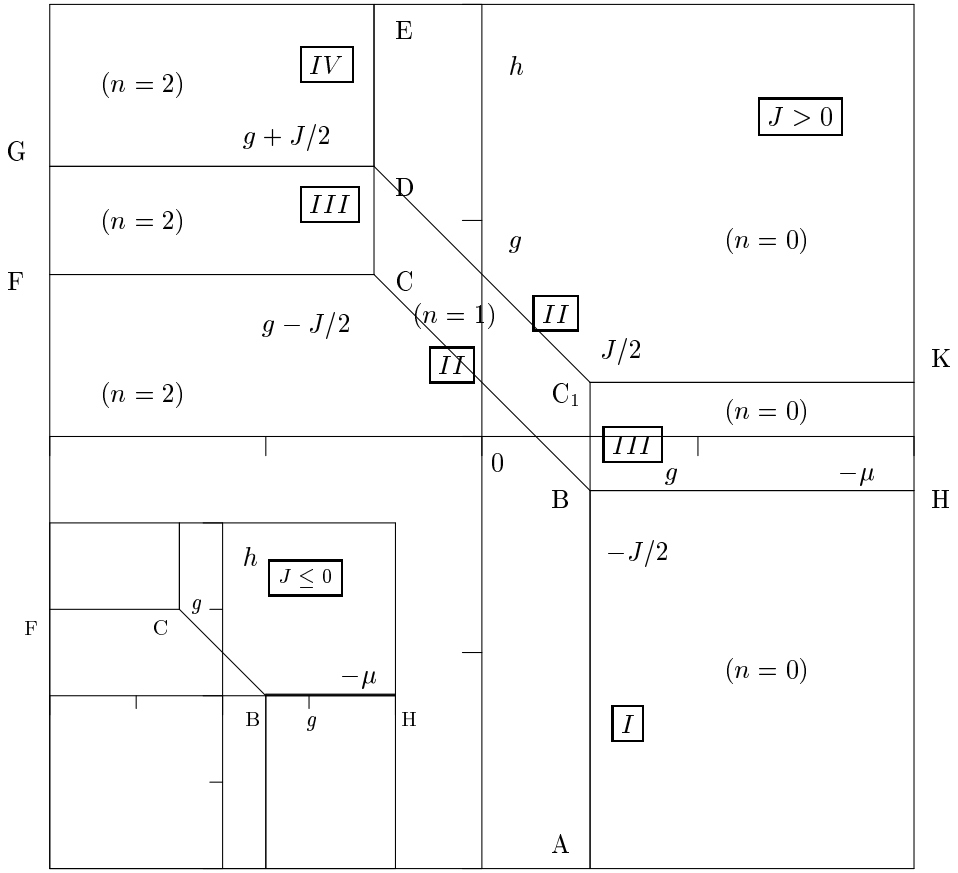


Figure 1. The ground state diagram. Average concentration of electrons is shown in the brackets.

$$\mu = const$$

$$\begin{aligned}
\mu \gg g \qquad & \chi_\perp = \frac{\beta d_s^2}{v_c(1+e^{-\beta\frac{J}{2}}\mathrm{ch}\beta(g-h))};\\
|\mu| \gg g,\ \mu<0 \qquad & \chi_\perp = \frac{\beta d_s^2}{v_c(1+e^{-\beta\frac{J}{2}}\mathrm{ch}\beta h)};\\
\mu \sim 0 \qquad & \chi_\perp(-h \gg g) = \frac{\beta}{v_c} d_e^2 e^{-\beta\frac{g}{2}},\\
& \chi_\perp(h \gg g) = 4\frac{\beta}{v_c} d_e^2 e^{-\beta\frac{g}{2}},\\
& \chi_\perp(h \sim \frac{g}{2}) = \frac{\beta}{v_c}(d_s - d_e)^2.
\end{aligned} \tag{8}$$

The formulae (7), (8) represent the main dependencies in the limit of low temperatures and are obtained in the case of infinite Coulomb repulsion U.

The influence of tunnelling motion in anharmonic potential wells on the susceptibility $\chi_\perp$ is essential when Ω splits the states which give at low temperatures the main contribution to the polarization. This effect can be taken into account by the perturbation theory. In this case the temperature dependence of $\chi_\perp$ is transformed from Curie's law to a more complicated form. The perturbational approach gives the following results:

$$n = const$$

$$\begin{aligned}
\boxed{\mathrm{I}},\ \boxed{\mathrm{II}},\ \boxed{\mathrm{IV}} \qquad & \chi_\perp \text{ is the same as in } \Omega = 0 \text{ case};\\
\boxed{\mathrm{III}} \qquad & \chi_\perp(0<n<1) = \frac{\beta}{v_c} n(d_s - d_e)^2,\\
& \chi_\perp(1<n<2) = \frac{\beta}{v_c}(2-n)(d_s - d_e)^2;
\end{aligned} \tag{9}$$

$$\mu = const$$

$$\mu \gg g \quad \chi_\perp = \frac{d_s^2}{v_c|\zeta_1+\zeta_2|} \times \frac{\mathrm{sh}\beta|\zeta_1+\zeta_2|}{\mathrm{ch}\beta(\zeta_1+\zeta_2) + e^{-\beta\frac{J}{2}}\mathrm{ch}\beta(g-h+\zeta_1-\zeta_2)},$$

$$\text{where } \zeta_1 = \frac{\Omega^2/4}{\frac{J}{2}+g-h},\ \zeta_2 = \frac{\Omega^2/4}{\frac{J}{2}-g+h};$$

$$-\mu \gg g \quad \chi_\perp = \frac{d_s^2}{v_c|\eta_1+\eta_2|} \times \frac{\mathrm{sh}\beta|\eta_1+\eta_2|}{\mathrm{ch}\beta(\eta_1+\eta_2)+e^{-\beta\frac{J}{2}}\mathrm{ch}\beta(h+\eta_1-\eta_2)},$$
$$\text{where } \eta_1 = \frac{\Omega^2/4}{\frac{J}{2}+h},\ \eta_2 = \frac{\Omega^2/4}{\frac{J}{2}-h}; \qquad (10)$$
$$\mu \sim 0 \quad \chi_\perp \text{ is the same as in } \Omega = 0 \text{ case.}$$

The tunnelling lowers the value of the transverse dielectric susceptibility $\chi_\perp$ almost in all range of parameters. When $\mu \to \pm\infty$ (the case when the influence of electron subsystem is absent), the susceptibility achieves maximum at low temperatures: from formula (10) we can obtain for susceptibility expression $\chi_\perp \overset{T\to 0}{\sim} \frac{d_s^2}{\Omega^2}$. In the region BCDC_1 the character of temperature dependence of the susceptibility is not changed. Here the tunnelling only shifts the states which contribute to the polarizability.

4. The mean field approximation.

The pseudospin part of the Hamiltonian $H_s + H_{s-s}$, except for the term containing J, corresponds to the Hamiltonian of Mitsui model. Model of this type is devoted to describe ferroelectrics with hydrogen bonds [12]. It also was proposed for the description of properties of superconductors of YBaCuO type [11]. We consider a simplified model and assume below that $\Omega = 0$, $U \to \infty$.

We consider the diagrammatic approach to derive the equations of the mean field approximation. Let us rewrite Hamiltonian (1) as follows:

$$\begin{aligned} H &= H_o - H_{int} \qquad (11)\\ H_o &= -\mu\sum_{i,s}(\hat{n}^s_{i1}+\hat{n}^s_{i2}) + U\sum_i(\hat{n}^\uparrow_{i1}\hat{n}^\downarrow_{i1}+\hat{n}^\uparrow_{i2}\hat{n}^\downarrow_{i2}) - J\sum_i S^z_{i1}S^z_{i2} + \\ &\quad +g\sum_{i,s}(\hat{n}^s_{i1}S^z_{i1}-\hat{n}^s_{i2}S^z_{i2}) - \sum_i(h_1S^z_{i1}+h_2S^z_{i2}) \\ h_1 &= h; \quad h_2 = -h \\ H_{int} &= \frac{1}{2}\sum_{i,i'}\sum_{\alpha,\beta}J^{\alpha\beta}_{ii'}S^z_{i\alpha}S^z_{i'\beta}. \end{aligned}$$

We introduce the Heisenberg representation of an operator in the second quantization form

$$\tilde{A}_i(\tau) = \mathrm{e}^{\tau H}A_i\mathrm{e}^{-\tau H}$$

and the interaction representation of this operator:

$$\tilde{A}_i(\tau) = \mathrm{e}^{\tau H_o}A_i\mathrm{e}^{-\tau H_o}.$$

Then the average of T-product $\langle T\,\tilde{A}_1(\tau_1)\dots\tilde{A}_n(\tau_n)\rangle$ over the Gibbs distribution with the full Hamiltonian may be written

$$\begin{aligned} \langle T\,\tilde{A}_1(\tau_1)\dots\tilde{A}_n(\tau_n)\rangle &= \frac{\langle T\,A_1(\tau_1)\dots A_n(\tau_n)S(\beta)\rangle_o}{\langle S(\beta)\rangle_o} \\ &= \langle T\,A_1(\tau_1)\dots A_n(\tau_n)S(\beta)\rangle^c_o, \qquad (12) \end{aligned}$$

where averaging $\langle\dots\rangle_o$ is carried out with the Hamiltonian H_o and will be treated according to diagrammatic technique procedure ($\langle\dots\rangle^c_o$ means that only connected diagrams will be taken into account), $\beta = 1/T$, and temperature S-matrix has the following form:

$$S(\beta) = \sum_{m=0}^{\infty}\frac{1}{m!}\int_0^\beta \mathrm{d}\tau_1\dots\int_0^\beta \mathrm{d}\tau_m T\{H_{int}(\tau_1)\dots H_{int}(\tau_m)\} \qquad (13)$$

Expression (12) is the basic one in the diagrammatic technique and allow us to find correlation functions and average values of operators.

We write the free energy in the zero approximation ($H_{int} = 0$):

$$\begin{aligned} F_o(h_\gamma) &= \mu n - \frac{T}{N}\mathrm{lnSp}(\mathrm{e}^{-\beta H_o}) = \qquad (14)\\ &= \mu n - T\ln\Big[2\Big\{e^{\beta\frac{J}{4}}\mathrm{ch}\frac{\beta}{2}(h_1+h_2)+e^{-\beta\frac{J}{4}}\mathrm{ch}\frac{\beta}{2}(h_1-h_2)\Big\} + \\ &+ 8e^{\beta\mu}\Big\{e^{\beta\frac{J}{4}}\mathrm{ch}\frac{\beta}{2}(h_1+h_2)\,\mathrm{ch}\frac{\beta}{2}g + e^{-\beta\frac{J}{4}}\mathrm{ch}\frac{\beta}{2}(h_1-h_2-g)\Big\} + \\ &+ 8e^{2\beta\mu}\Big\{e^{\beta\frac{J}{4}}\mathrm{ch}\frac{\beta}{2}(h_1+h_2)+e^{-\beta\frac{J}{4}}\mathrm{ch}\frac{\beta}{2}(h_1-h_2-2g)\Big\}\Big] \end{aligned}$$

(we show explicit dependence of the free energy from field h_γ, $\gamma = 1,2$ – coefficient near $S^z_{i\gamma}$). Then the general expression for free energy is

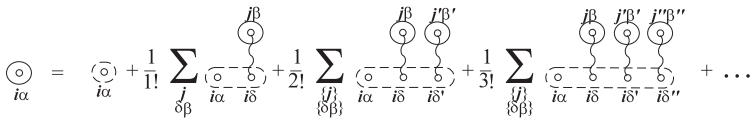
$$\begin{aligned} F &= \mu n - \frac{T}{N}\mathrm{lnSp}(\mathrm{e}^{-\beta H}) = F_o(h_\gamma) - \frac{T}{N}\ln\langle S(\beta)\rangle_o \\ &= F_o(h_\gamma) - \frac{T}{N}\langle S(\beta)\rangle^c_o. \qquad (15) \end{aligned}$$

We introduce the following diagrammatic representation:

$= J^{\alpha\beta}_{ij}$

$$\begin{aligned} \circ_{i\alpha} &= \langle S^z_{i\alpha}\rangle_o = -\frac{\partial}{\partial\ h_\alpha}F_o(h_\gamma) = b_\alpha(h_\gamma) \\ \circ\!\!-_{i\alpha} &= \langle S^z_{i\alpha}\rangle \\ \circ\!\!-\!\!-\circ &= b_{\alpha\alpha'}(h_\gamma) \equiv b_{\alpha\alpha'} = \frac{\partial}{\partial\beta h_{\alpha'}} b_\alpha(h_\gamma) \end{aligned}$$

The summation of the one-tail diagrams corresponds to the mean field approximation:



(16)

or in terms of formulae:

$$\begin{aligned} \langle S^z_\alpha\rangle &= b_\alpha(h_\gamma) + \frac{\beta}{1!}\sum_{\delta\beta}\sum_j J^{\delta\beta}_{ij}\langle S^z_\beta\rangle \frac{\partial}{\partial\beta h_\delta} b_\alpha(h_\gamma) + \qquad (17) \\ &+\frac{\beta^2}{2!}\sum_{\delta\delta'\beta\beta'}\sum_{jj'} J^{\delta\beta}_{ij}\langle S^z_\beta\rangle J^{\delta'\beta'}_{ij'}\langle S^z_{\beta'}\rangle \frac{\partial^2}{\partial\beta h_{\delta'}\partial\beta h_\delta} b_\alpha(h_\gamma) + \ldots . \end{aligned}$$

Noticing that the right side of this relation contains the usual expansion of $b_\alpha(h_\gamma)$ over parameter h_γ, we rewrite

$$\begin{aligned} \langle S^z_\alpha\rangle &= b_\alpha(h_\gamma + \sum_\delta \beta J_{\delta\gamma}\langle S^z_\delta\rangle) \equiv b_\alpha(\tilde{h}_\gamma), \text{ where} \qquad (18) \\ J_{\delta\gamma} &\equiv J_{\delta\gamma}(\mathbf{k}=0) = \sum_j J^{\delta\gamma}_{ij} \end{aligned}$$

Formula (18) is the transcendental system of equations ($\alpha = 1, 2$) from which one can determine the polarization $\eta = \langle S^z_1 + S^z_2\rangle$ in the molecular field approximation. In the case of constant concentration we need to supplement the system (18) by the equation which defines the chemical potential

$$\frac{\mathrm{Sp}\left(\hat{n}\mathrm{e}^{-\beta(H_o - \sum_{\delta\gamma} J_{\delta\gamma}\langle S^z_\delta\rangle S^z_\gamma)}\right)}{\mathrm{Sp}\left(\mathrm{e}^{-\beta(H_o - \sum_{\delta\gamma} J_{\delta\gamma}\langle S^z_\delta\rangle S^z_\gamma)}\right)} = n$$

In the mean field approximation the free energy has the following form:

$$F_{MFA} = \mu n + \frac{\beta}{2}\sum_{\delta\gamma} J_{\delta\gamma}\langle S^z_\delta\rangle\langle S^z_\gamma\rangle - \frac{T}{N}\ln \mathrm{Sp}\ \mathrm{e}^{-\beta(H_o - \sum_{\delta\gamma} J_{\delta\gamma}\langle S^z_\delta\rangle S^z_\gamma)}$$

$$= F_o(\tilde{h}_\gamma) + \frac{\beta}{2}\sum_{\delta\gamma} J_{\delta\gamma}\langle S^z_\delta\rangle\langle S^z_\gamma\rangle \qquad (19)$$

The phase diagrams which describe the ferroelectric type phase transitions for cases $\mu = const$ and $n = const$ are shown on Fig.2-3 (all model parameters are derived by $J_{11} + J_{12} > 0$). It may be easily seen that in case of $\frac{J_{11}-J_{12}}{J_{11}+J_{12}} = -1$ (only interplane interaction is present) exclusively the structural phase transition of second order takes place. Increasing of parameter $\frac{J_{11}-J_{12}}{J_{11}+J_{12}}$ leads to narrowing of the ferroelectric ordering area and appearance of the first order phase transition. The width of this area is proportional to J in $\mu = const$ and may disappear if $J = 0$. This points to the importance of explicit allowing for the interaction J within cluster.

In $n = const$ regime ferroelectric phase exists at all values of J. This is explained by peculiarities of behaviour of the chemical potential in this case. Coming back to the ground state diagram (Fig.1) we note that the chemical potential varies along curve AC_1DE at $n \sim 0$ and along ABCD at $n \sim 2$. The area of degenerated ground state (in which there is always ferroelectric ordering) is the parallelogram $BCDC_1$ which length along axis $0h$ is $g + J$. This value defines approximate width of ferroelectric phase area.

The presence of a phase transitions manifests in the peculiar dependencies of dielectric susceptibility on h ([14]). In the vicinity of a second order phase transition point the susceptibility increases drastically. At the point of first order phase transition the susceptibility jumps to another value without singularities.

The long-range interaction eliminates the $\frac{1}{T}$ law. It may be shown easy in the $\mu = const$ regime. We can neglect the field dependence of parameters S^z_α ($\eta = \langle S^z_1 + S^z_2\rangle = 1$ and $\xi = \langle S^z_1 - S^z_2\rangle = 0$ in pseudospin-ordered phase) and use the formula

$$\chi_\perp = \frac{\beta}{v_c}\langle (P^z)^2\rangle - \frac{\beta}{v_c}\langle P^z\rangle^2.$$

$\langle P^z\rangle$ is different from zero in the ferroelectric phase. Moreover, $\langle (P^z)^2\rangle \stackrel{T\to 0}{=} \langle P^z\rangle^2$. In the limit $\mu \to -\infty$, for example,

$$\chi_\perp = \frac{\beta}{v_c} 4 d_s^2 e^{-\beta|J_{11}+J_{12}|} \qquad (20)$$

in the ferroelectric phase ($|h| < \frac{J}{2}$, $J > 0$). The temperature behaviour

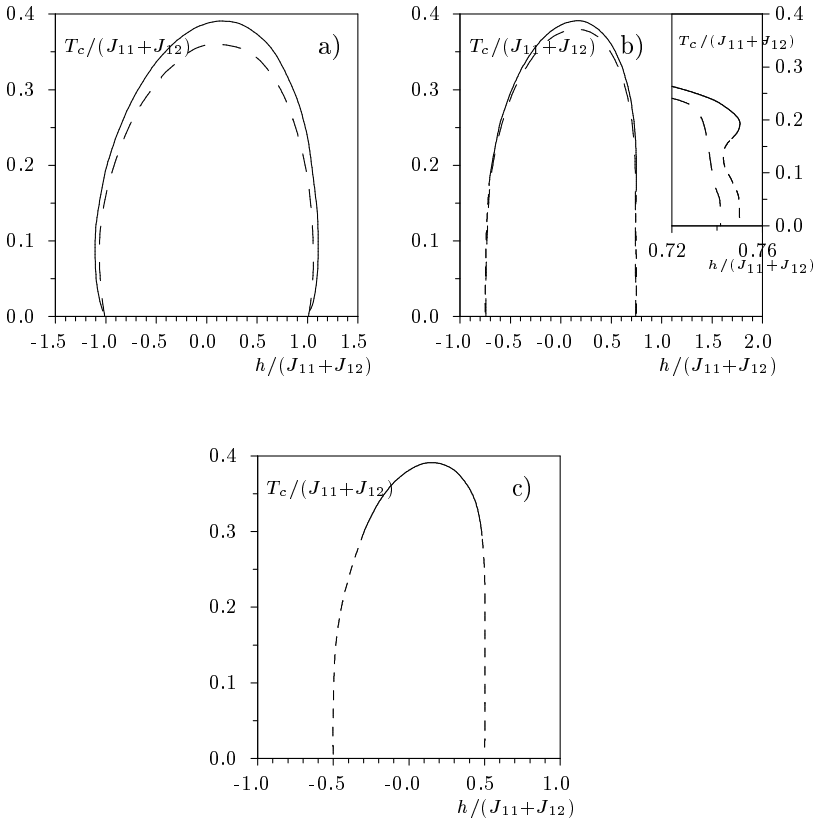


Figure 2. Dependence of the temperature of ferroelectric phase transition T_c on h at different values of parameter $J_{11}-J_{12}$ in the regime $\mu = const$: a) $\frac{J_{11}-J_{12}}{J_{11}+J_{12}} = -1$, b) $\frac{J_{11}-J_{12}}{J_{11}+J_{12}} = 0$, c) $\frac{J_{11}-J_{12}}{J_{11}+J_{12}} = 1$. Other parameters: $J/(J_{11}+J_{12}) = 1$, $g/(J_{11}+J_{12}) = 1$, $\mu/(J_{11}+J_{12}) = -1$. The phase transitions can be of second (solid lines) or of first order (dashed lines). The widely spaced dashed lines correspond to one-loop approximation with $N_{int} = 2$.

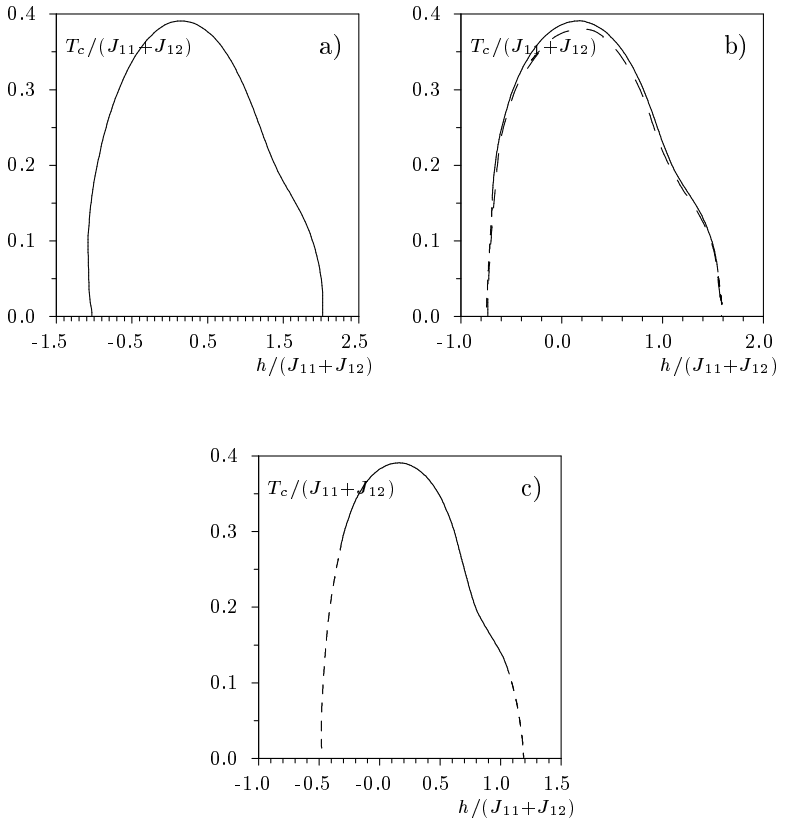


Figure 3. h dependence of the temperature of ferroelectric phase transition T_c at different values of parameter $J_{11}-J_{12}$ in the regime $n = const$: a) $\frac{J_{11}-J_{12}}{J_{11}+J_{12}} = -1$, b) $\frac{J_{11}-J_{12}}{J_{11}+J_{12}} = 0$, c) $\frac{J_{11}-J_{12}}{J_{11}+J_{12}} = 1$. Other parameters: $J/(J_{11}+J_{12}) = 1$, $g/(J_{11}+J_{12}) = 1$ $n = 0.4$. Solid lines and dashed lines represent second order and first order phase transitions respectively. The widely spaced dashed line corresponds to one-loop approximation with $N_{int} = 2$.

of susceptibility is changed also in the paraphase ($|h| > \frac{J}{2}$, $J > 0$):

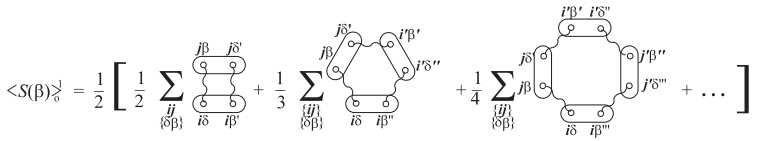
$$\chi_\perp = \frac{\beta d_s^2}{v_c\left(1 + e^{-\beta\frac{J}{2}}\mathrm{ch}\beta(\frac{J_{11}-J_{12}}{2} + |h|)\right)}. \qquad (21)$$

The last expression at $J_{11} = J_{12} = 0$ agrees with the result obtained in the case of the absence of a long-range interaction (8).

5. One-loop approximation.

In constructing of higher order approximation we can use mean field approximation as the zero one. Then it is assumed that all one-tail diagrams are already summarized and all averaging is carried out over distribution with the Hamiltonian $H_{MFA} = H_o - \sum_{\delta\gamma} J_{\delta\gamma}\langle S_\delta^z\rangle S_\gamma^z + \frac{1}{2}\sum_{\delta\gamma} J_{\delta\gamma}\langle S_\delta^z\rangle\langle S_\gamma^z\rangle$. We represent this graphically by substituting blocks surrounded by dashed lines into blocks surrounded by solid line. In one-loop approximation the free energy has the form

$$F^1 = F_{MFA} - \frac{T}{N}\langle S(\beta)\rangle_o^1 \qquad (22)$$



Introducing matrixes $\hat{\mathbf{b}}$ and $\hat{\mathbf{J}}$ with elements $b_{\alpha\beta}$ and $J_{\alpha\beta}$ correspondingly and making Fourier transformation, we rewrite (22) in the following form:

$$\begin{aligned}\langle S(\beta)\rangle_o^1 &= \frac{1}{2}\left[\sum_{\mathbf{k}} \mathrm{Sp}\left\{\frac{1}{2}(\hat{\mathbf{J}}(\mathbf{k})\hat{\mathbf{b}})^2 + \frac{1}{3}(\hat{\mathbf{J}}(\mathbf{k})\hat{\mathbf{b}})^3 + \frac{1}{4}(\hat{\mathbf{J}}(\mathbf{k})\hat{\mathbf{b}})^4 + \dots\right\}\right] \\ &= -\frac{1}{2}\{\sum_{\mathbf{k}}\sum_{i=1}^{2} \ln(1-\lambda_i(\mathbf{k}))\}, \qquad (23)\end{aligned}$$

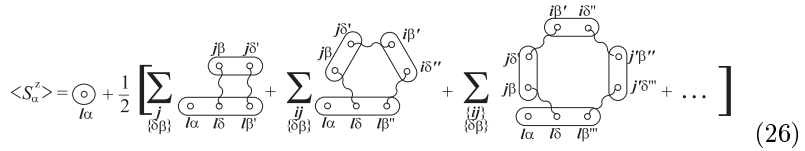
where $\lambda_i(\mathbf{k})$ are the eigenvalues of $\hat{\mathbf{J}}(\mathbf{k})\hat{\mathbf{b}}$ matrix:

$$\lambda_{1,2}(\mathbf{k}) = \frac{J_{11}(\mathbf{k})(b_{11}+b_{22}) + 2J_{12}(\mathbf{k})b_{12}}{2} \pm \qquad (24)$$
$$\pm\sqrt{\frac{J_{11}^2(\mathbf{k})(b_{11}-b_{22})^2}{4} + (J_{11}(\mathbf{k})b_{12} + J_{12}(\mathbf{k})b_{11})(J_{11}(\mathbf{k})b_{12} + J_{12}(\mathbf{k})b_{22})}$$

The equation for $\langle S_\alpha^z\rangle$ can be easily derived by making the derivation of free energy of one site over the $\tilde{h}_\alpha$:

$$\langle S_\alpha^z\rangle = -\frac{\partial F^1}{\partial\tilde{h}_\alpha} = b_\alpha - \frac{T}{2N}\sum_{\mathbf{k}}\sum_{i=1}^{2}\frac{1}{1-\lambda_i(\mathbf{k})}\frac{\partial\lambda_i(\mathbf{k})}{\partial\beta\tilde{h}_\alpha} \qquad (25)$$

The corresponding diagrammatic series for the average value of pseudospin has the form



(26)

Summing over $\mathbf{k}$ which is contained in formulas (23) and (25) in this approximation is usually performed by introducing the density of states (which is rectangular in two dimensional case). The final expression for free energy is

$$F^1 = F_{MFA} - \frac{1}{\beta} - \frac{1}{4\beta}\sum_{i=1}^{2}\left\{\frac{1}{\beta\lambda_i(0)}\ln\frac{1-\beta\lambda_i(0)}{1+\beta\lambda_i(0)} - \ln(1-\beta^2\lambda_i(0)^2)\right\},$$
$$\lambda_i(0) \equiv \lambda_i(\mathbf{k}=0) \quad \text{for} \quad i = 1,2. \qquad (27)$$

The solution of system (25) in the particular case of second order phase transition at $\mu = const$ is shown on Fig.4. The temperature of phase transition to the ferroelectric phase T_c is lower if compared with the one derived in the mean field approximation. However, in this approach one-loop approximation points to the possibility of appearing non-physical phase transitions inside ordered phase. In [15] where the spinless fermion model was considered in order to avoid undesirable phase transitions it is proposed to apply the correctional high-density expansion (CHDE) method or to renormalize the semi-invariants by summing "two-tailed" parts (this scheme takes into account gaussian fluctuations of molecular field). From our point of view, additional phase transitions are not the defect of the approximation but are the result of its unjustified application for short-range interaction. The Fourier image of the potential of nearest neighbours has the form

$$J(\mathbf{k}) = \frac{J}{2}(\cos k_x a + \cos k_y a), \qquad (28)$$

where a is the lattice constant. Indeed, the one-loop approximation takes into account contributions of the diagrams with one sum over $\mathbf{k}$. According to classification given in [13] it corresponds to correction of the first

order in $(a/R_o)^2$ expansion (R_o is the radius of interaction). That is why we can not use (28) for one-loop approximation.

It is well-known that the mean field approximation gives the exact result in case when the interaction between the sites has infinite radius and does not depend on the distance:

$$J_{ij} = \frac{J}{N} - \frac{J}{N}\delta_{ij}, \tag{29}$$

or, after Fourier transformation,

$$J(\mathbf{k}) = J\delta_{\mathbf{k}=0} - \frac{J}{N}. \tag{30}$$

If we use this potential in the expressions for elementary one-loop diagrams, the correction to the free energy and average values of pseudospins is proportional to $\frac{1}{N}$ and disappears in the thermodynamical limit $N \to \infty$. When passing from infinitely long interaction to the interaction with definite radius $J(\mathbf{r}, R_o)$, we should satisfy the sum rule for Fourier image (which exclude self-interaction)

$$\sum_{\mathbf{k}} J(\mathbf{k}) = 0. \tag{31}$$

and $J(\mathbf{k})$ should corresponds (30) in the extreme case $R_o \to \infty$. Let us define the potential of interaction $J(\mathbf{r}, R_o)$ in the form

$$J_{\delta\gamma}(\mathbf{r}, R_o) = \frac{J_{\delta\gamma}}{2\pi}\frac{a^2}{R_o^2}\mathrm{e}^{-r/R_o}, \tag{32}$$

which is widely applied to describe the interactions in spin models. Taking into account the condition (31), we can write its Fourier image in the form

$$J_{\delta\gamma}(\mathbf{k}) = \frac{J_{\delta\gamma}}{(1+k^2R_o^2)^{\frac{3}{2}}} - \underbrace{\frac{1}{N}\sum_{\mathbf{k}}\frac{J_{\delta\gamma}}{(1+k^2R_o^2)^{\frac{3}{2}}}}_{C}, \tag{33}$$

or, using discrete representation for $\mathbf{k}$ vector,

$$J_{\delta\gamma}(m,n) = \frac{J_{\delta\gamma}}{(1+4\pi^2(n^2+m^2)N_{int}^2/N)^{\frac{3}{2}}} - C, \tag{34}$$

where $n, m = -\sqrt{N}+1, \ldots, 0, 1, 2, 3, \ldots, \sqrt{N}$, $N_{int} = R_o/a$ in the numerical calculations. We use the simplest method taking the direct summing over $\mathbf{k}$, choosing $\sqrt{N} = 40$, $N_{int} = 2$ (number of neighbours which

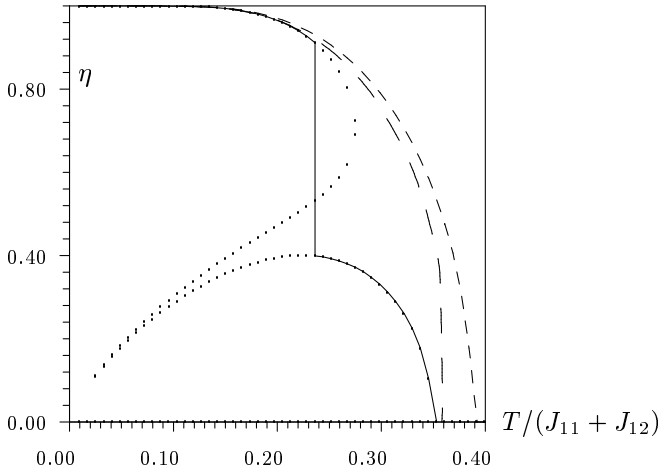


Figure 4. The order parameter $\eta = \langle S_1^z + S_2^z \rangle$ vs temperature T in different approaches: dashed line – MFA, solid line – one-loop approximation with using rectangular density of states which corresponds to the short-range interaction, widely spaced dashed line – one-loop approximation with direct summing over Brillouin zone ($N = 1600$, $N_{int} = 2$). $\mu/(J_{11}+J_{12}) = -1$, $\frac{J_{11}-J_{12}}{J_{11}+J_{12}} = -1$, $J/(J_{11}+J_{12}) = 1$, $g/(J_{11}+J_{12}) = 1$.

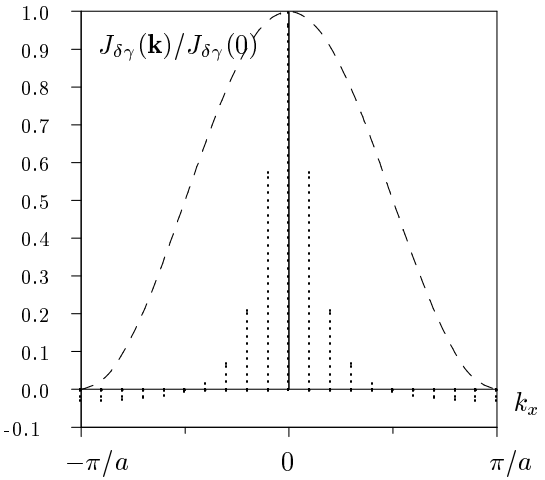


Figure 5. The dependence of $J(\mathbf{k})$ on k_x at $k_y = 0$. Solid line represents long range interaction, dashed line - nearest neighbour approximation, dotted line corresponds to e^{-r/R_o} potential.

interact with a given site is ~ 16, thus having long-range interaction). $J(\mathbf{k}) \to 0$ at the border of Brillouin zone, therefore the condition of periodicity of $J(\mathbf{k})$ in the reverse space is valid. The results of numerical calculations of the order parameter and phase diagrams are shown on Fig.4. As it may be seen, the temperature of the ferroelectric phase transition became lower, although not very much (up to 10% of the temperature magnitude due to a large number of interacting neighbours). As it seen from Fig.2, existence of narrow interval of the asymmetry parameter h value, at which three phase transitions exist is the result of long-range forces. This feature disappears when radius of interaction became smaller than some definite value.

It may be easily seen that width of the ferroelectric phase stays practically unchanged and conclusions which were made basing on the results of molecular field are valid for the higher approximation. The same may be concluded about the type of phase transition.

6. Conclusions.

In the framework of two-sublattice pseudospin-electron model the transverse dielectric susceptibility $\chi_\perp$ is calculated in the case of the absence of electron transfer. It is shown that if $J > 0$ (J describes the interaction between pseudospins in the same cell), there is a region of values of h parameter with width $\sim J$ where the ground state of the system is degenerate. Here, in the regime $\mu = const$, susceptibility is inversely proportional at low temperatures to the temperature. By contrast, in the regime $n = const$ ($\mu = \mu(n)$) the dielectric susceptibility obeys Curie's law at all values of the model parameters. The role of tunnelling is significant in the region where Ω splits the degenerate ground state. Then the susceptibility achieves its maximum at $T = 0$ ($\chi_\perp \sim \frac{1}{\Omega^2}$).

In the second part of this work the interaction between pseudospins was considered within the mean field approximation as well as one-loop approximation. In substance, we dealt with a Mitsui model supplemented by an interaction with an electron system. On the basis of this model the ferroelectric type instabilities and phase transitions, connected with macroscopic polarization along c-axis in the systems like YBaCuO are considered. It should be noted that the problem of the existence of state with spontaneous polarization has been subject for study for recent years. It was reported, for example, about the observation of pyroelectric properties of YBaCuO [10], [16].

The interaction between electron and pseudospin subsystems leads to a shift of the ferroelectric phase compared to the pure Mitsui model

and to a change in shape of the phase diagrams. The ferroelectric phase is always present in the region with degenerate ground state. The width of the corresponding interval of values of h parameter is proportional to the constant of interaction between pseudospins inside cluster. The appearance of a pseudospin-ordered phase changes the temperature behaviour of $\chi_\perp$ near phase transition points and eliminates Curie's law in the ferroelectric phase.

The analysis of the model in the approximation higher than the mean field, namely in the so-called one-loop approximation was carried out. It was shown that the right choice of the model long-range potential excludes additional non-physical phase transitions in the ordered phases typical for this approximation. The lowering of the temperature of transition from paraphase to to ferroelectric phase was established, as well as the narrowing of the ferroelectric ordering area. The existence of the ranges of h-parameter (both in this and in Mitsui model) at which three phase transitions exist is the result related to the long-range interaction between pseudospins.

It should be mentioned that ferroelectric ordering of pseudospins has an effect on the redistribution of the electrons (holes) between superconducting planes. On the other hand, electron transfer in the superconducting planes produces an effective interaction between pseudospins [8,9]. That is why it is necessary to consider both processes self-consistently to describe the phase transitions and instabilities in real systems.

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Структурні фазові переходи в двопідгратковій псевдоспін-електронній моделі високотемпературних надпровідників

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