

COUPLED BIPOLARONS AND OPTICAL PHONONS AS A MODEL FOR HIGH- T_c SUPERCONDUCTORS

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1. INTRODUCTION.

All known up-to-date high-temperature superconductors are oxides (mainly copper oxides although compounds without copper were obtained, e.g. $BaKBiO_3$).

On the other hand, more than 95 % of all ferroelectric compounds are also oxides or contain oxygen ion [1] . Therefore, the problem of mutual relation between ferroelectricity and superconductivity is of great theoretical interest.

The electron-phonon interaction is important in both phenomena. In ferroelectrics a cubic electron-phonon and/or a quartic electron-two-phonon interactions play an essential role [1] . In classical superconductors (of the BCS type) a cubic electron-one-phonon interaction leads to the formation of Cooper pairs of two electrons with opposite momenta and spins . The average distance (in real space) between the two electrons is of order of the so-called coherence length ξ which is much greater than lattice constant of such a classical superconductor. However, the coherence length of the new high-temperature superconductors reaches very small value which is comparable to the dimensions of unit cell of these compounds [3] . This means that a pair consists of two holes occupying the same site or two adjacent sites . Such a situation seems to be described by a model of the local-pairs (bipolarons) [2,3].

The origin of local-pair may come not only from strong enough electron or hole-phonon interaction but also from other interactions . Independently of a specific nature of such local-pairs, they can undergo a Bose-like condensation to the superconducting state at a critical temperature which is usually much lower than the temperature of the pair formation.

In this paper an interplay of ferroelectric and superconducting properties is considered within the model of hole-like local-pairs interacting with optical phonons . Therefore, we extend the usual local-pair Hamiltonian [2] by including an direct interaction between the local-pairs and the optical phonons. These optical phonons are known to play an important role in the ferroelectric transition if any and they transform into an additional pseudo-acoustic branch at the ferroelectric critical temperature [1] (this is associated with arising of nonzero electric polarization due to existence of two separate lattices composed of negative and positive ions, respectively).

2. HAMILTONIAN OF INTERACTING LOCAL-PAIRS AND OPTICAL PHONONS.

The Hamiltonian of our system is as follows

$$H = -w_0 \sum_i \sum_j (N_i N_j - A_i^\dagger A_j) + \sum_i (\hbar\nu_0 b_i^\dagger b_i - \mu N_i - \lambda N_i (b_i^\dagger + b_i)), \text{ where}$$

$A_i^\dagger(A_i), b_i^\dagger(b_i)$ are creation (annihilation) operators for the local-pair and the optical phonon (with frequency ν_0) at the i -th site, respectively, $N_i = A_i^\dagger A_i$, w_0 is a local-pair bandwidth and μ denotes the chemical potential. An analysis

of the above Hamiltonian can be carried out on the ground of the Bogolyubov's inequality [4] for the free energy $F \leq F_t = F_0 + \langle H \rangle_0 - \langle H_t \rangle_0$, with the trial Hamiltonian in which the local-pairs and the phonons are decoupled

$$H_t = \sum_i (-W_i A_i^\dagger - W_i^* A_i - Q_i N_i + \hbar \nu_0 B_i^\dagger B_i) = H_{\text{pair}} + H_{\text{ph}}$$

where W_i , W_i^* and Q_i stand for variational parameters and B_i^\dagger (B_i) are new creation (annihilation) operators for phonon at the i -th site. The thermal averages of the $\langle \dots \rangle_0$ type are defined as usual, e.g., $\langle H \rangle_0 = \text{Tr} (e_0 H)$, $e_0 = \exp(-\beta H_t) / \text{Tr} (\exp(-\beta H_t)$, $\beta = (k_B T)^{-1}$, k_B - the Boltzmann's constant. Within the single site approximation which is equivalent to the mean-field approximation (MFA)

$$F_0 = -\beta^{-1} \sum_i \ln Z_0 \quad \text{with the single-site partition function}$$

$$Z_0 = (\exp(-\beta \varepsilon_1) + \exp(-\beta \varepsilon_2) / (1 - \exp(-\beta \hbar \nu_0))) ,$$

where ε_1 and ε_2 are the eigenvalues of the Hamiltonian H_{pair} calculated in the basis of states consisting of two single-site states: φ_0 and φ_1 which describe a given site occupied (φ_1) or unoccupied (φ_0) by a local-pair. A

straightforward diagonalization of the H_{pair} matrix $\begin{pmatrix} 0 & -W^* \\ -W & Q \end{pmatrix}$ leads to the following eigen-energies

$$\varepsilon_1 = (-Q - (-Q^2 + 4|W|^2)^{1/2}) / 2$$

$$\varepsilon_2 = (-Q + (-Q^2 + 4|W|^2)^{1/2}) / 2$$

The variational parameters Q , W and W^* should be calculated by minimizing the trial free energy F_t . One obtains the mean-field Hamiltonian H_{MFA} by means of the following decouplings

$$\sum_i \sum_j A_i^\dagger A_j \Rightarrow \sum_i \sum_j \langle A_i^\dagger \rangle A_j + \sum_i \sum_j A_i^\dagger \langle A_j \rangle = \sum_i z (\alpha A^\dagger + \alpha^* A)$$

$$\sum_i \sum_j N_i N_j \Rightarrow \sum_i \sum_j \langle N_i \rangle N_j + \sum_i \sum_j N_i \langle N_j \rangle = 2z \nu \sum_i N_i$$

where $\alpha = \alpha_i = \langle A_i \rangle = \langle A \rangle$ is the superconducting order parameter, $\nu = \nu_i = \langle N_i \rangle = \langle N \rangle$ is the concentration of the local-pairs per one site and z denotes a coordination number (a number of the nearest neighbours of a given site). In the result

$$H_{\text{MFA}} = \sum_i (-w (\alpha A^\dagger - \alpha^* A) + 2w \nu N + \hbar \nu_0 b^\dagger b - \mu N - \lambda N (b^\dagger + b))$$

with the parameter $w = z w_0$.

3. FREE ENERGY AND COUPLING EQUATIONS.

In this section we determine the trial free energy according to the

above Hamiltonian. The trial energy per one site $f_t = F_t / \sum 1$ takes the form

$$f_t = -\beta^{-1} \ln(\exp(-\beta \varepsilon_1) + \exp(-\beta \varepsilon_2)) + \beta^{-1} \ln(1 - \exp(-\beta \hbar \nu_0)) + W \alpha^* + W^* \alpha - 2w|\alpha|^2 + 2w^2 + (Q-\mu)\nu + \hbar \nu_0 (\langle b^\dagger b \rangle_0 - \langle B^\dagger B \rangle_0) - \lambda \langle N(b^\dagger + b) \rangle_0$$

In order to calculate the necessary averages let us introduce the new states

$$\Phi_1 = c_{10} \varphi_0 + c_{11} \varphi_1 \quad \Phi_2 = c_{20} \varphi_0 + c_{21} \varphi_1$$

for which the matrix of H_{pair} has a diagonal form. The coefficients c_{kl} can

be obtained from the equation: $H_{\text{pair}} \Phi_i = \varepsilon_i \Phi_i$, i.e.

$$\begin{pmatrix} -\varepsilon_i & -W^* \\ -W & Q - \varepsilon_i \end{pmatrix} \begin{pmatrix} c_{i0} \\ c_{i1} \end{pmatrix} = 0$$

Taking into account the normalization condition for eigenfunctions Φ_1 and Φ_2 one obtains in the result:

$$c_{i0} = (1 + |\varepsilon_i / W^*|^2)^{-1/2} = (W / (Q - \varepsilon_i)) (1 + |W^* / (Q - \varepsilon_i)|^2)^{-1/2}$$

$$c_{i1} = (1 + |W^* / (Q - \varepsilon_i)|^2)^{-1/2} = (-\varepsilon_i / W^*) (1 + |-\varepsilon_i / W^*|^2)^{-1/2}$$

Consider the quantities $\varepsilon_k = \langle \Psi_k | b - B | \Psi_k \rangle$, where Ψ_k are the phonon eigenfunctions

i.e. $H_{\text{ph}} \Psi_k = \hbar \nu_0 \Psi_k$ ($k = 0, 1, 2, \dots$). Let us assume for simplicity that

$\varepsilon_k = \varepsilon$ and $\langle \Psi_{k-1} | b - B | \Psi_k \rangle = 0$ for all values of k . The parameter ε can be

treated as a non-variational quantity which is proportional to a lattice deformation. This deformation can be associated with arising a nonzero dipole electric moment, i.e. with a ferroelectric phase transition. After some algebra one comes to the following:

$$\langle b^\dagger b \rangle_0 - \langle B^\dagger B \rangle_0 = \varepsilon^* \varepsilon.$$

Using the calculated coefficients c_{i1} one obtains:

$$\langle N b \rangle_0 = \varepsilon / 2 + \varepsilon Q (Q^2 + 4|W|^2)^{-1/2} \left[2 - \tanh(\beta(Q^2 + 4|W|^2)^{1/2}) \right] / 2$$

The trial reduced free energy is then readily written as

$$f_t = -\beta^{-1} \ln(\exp(-\beta \varepsilon_1) + \exp(-\beta \varepsilon_2)) + \beta^{-1} \ln(1 - \exp(-\beta \hbar \nu_0)) + W \alpha^* + W^* \alpha - 2w|\alpha|^2 + 2w^2 + (Q-\mu)\nu + \hbar \nu_0 \varepsilon^* \varepsilon - \lambda (\varepsilon^* + \varepsilon) \left[1 + Q (Q^2 + 4|W|^2)^{-1/2} \left[2 - \tanh(\beta(Q^2 + 4|W|^2)^{1/2}) \right] \right] / 2$$

A minimization of the above thermodynamic potential with respect to W , W^* , α , α^* , Q and ν leads to the following coupling equations

$$(2w)^{-1} = (Q^2 + 4|W|^2)^{-1/2} \tanh(\beta(Q^2 + 4|W|^2)^{1/2}) - \lambda (\varepsilon^* + \varepsilon) Q (Q^2 + 4|W|^2)^{-3/2}$$

$$\tanh(\beta(Q^2 + 4|W|^2)^{1/2}) + \lambda (\varepsilon^* + \varepsilon) \beta Q (Q^2 + 4|W|^2)^{-1} \left[\cosh(\beta(Q^2 + 4|W|^2)^{1/2}) \right]^{-2} / 2$$

$$2\nu - 1 = Q (Q^2 + 4|W|^2)^{-1/2} \tanh(\beta(Q^2 + 4|W|^2)^{1/2}) + 4\lambda (\varepsilon^* + \varepsilon) |W|^2 (Q^2 + 4|W|^2)^{-3/2}$$

$$\tanh(\beta(Q^2 + 4|W|^2)^{1/2}) + \lambda (\varepsilon^* + \varepsilon) \beta Q^2 (Q^2 + 4|W|^2)^{-1} \left[\cosh(\beta(Q^2 + 4|W|^2)^{1/2}) \right]^{-2} / 2$$

$$W = 2w\alpha \quad Q = -4w\nu + \mu \quad + \text{c.c.}$$

It is easily seen that $W = W^*$ and $\alpha = \alpha^*$. The above set of equation should be

self-consistently solved.

4. GROUND STATE .

By taking the limit $T \rightarrow 0$, the energy of a ground state per one site is given by

$$E = \langle H \rangle_0 = \sum_i 1 = -2w\alpha^2 + 2w\nu^2 - \mu\nu + \hbar\nu_0 \xi^2 + \lambda\xi(1+Q/(Q^2+4W^2)^{1/2})$$

Let us consider the purely superconducting ground state for which the ferroelectric order parameter ξ disappears ($\xi=0$). The superconducting order parameter strongly depends on the local-pair concentration, i.e. $\alpha^2 = \nu(1-\nu)$. The energy of the superconducting ground state is as follows

$$E_{sc} = -2w\alpha^2 + 2w\nu^2 = 2w(2\nu-1)\nu$$

For the purely ferroelectric ground state the superconducting order parameter disappears ($\alpha=W=0$) and the energy is given by

$$E_{fe} = 2w\nu^2 + \hbar\nu_0 \xi^2 - \lambda\xi(1+Q/(Q^2+4W^2)^{1/2})$$

The normal state, i.e. non-superconducting and paraelectric ($\alpha=0$ and $\xi=0$), has the energy $E_n = 2w\nu^2$. It is clearly seen that normal state cannot be realized because of the lower energy of the superconducting state, at least ($E_{sc} < E_n$). Nevertheless, a competition is possible between superconducting and ferroelectric ground states. The superconducting state is preferred if $2\nu(1-\nu) > (2\lambda\xi - \hbar\nu_0 \xi^2)/w$.

5. SUPERCONDUCTING TRANSITION

Let us return to finite temperatures. To get the superconducting critical temperature T_c we assume that superconducting order parameter disappears ($\alpha=\langle A \rangle=0$).^c The reduced critical temperature is thus given by

$$t_c = k_B T_c / w = q / \ln((1+Z)/(1-Z)),$$

where $q = Q/w = 2\nu-1+((2\nu-1)^2-4pZ)^{1/2}$, $p = \lambda \xi / w$ and the quantity Z can be numerically calculated from the additional condition

$$0 = q^2/2 - (q-2p)Z - p(1-Z^2) \ln((1+Z)/(1-Z))$$

It can be shown that maximum critical temperature is strongly enhanced due to the rather moderate interaction up to $p \approx 0.3$. However, a nonzero value of ξ , i.e. temperature below ferroelectric transition temperature, is necessary. For stronger coupling and/or smaller bandwidth w , this effect weakens. On the other hand, the high-temperature superconductivity is rather restricted to the regions far from half-filling ($\nu \neq 1/2$).

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