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COUPLED STATES IN ELECTRON-PHONON SYSTEM, SUPERCONDUCTIVITY AND HIGH T_c SUPERCONDUCTIVITY OF CRYSTALS

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The new theoretical approach is proposed for studying the coupled states responsible for superconductivity in crystal without using the idea of electron pairing. From our approach it follows that BCS electron-pairing postulates are only approximate. It is shown that superconductivity (SC) coupled states can be created by pairs of electrons with nonzero full momentum and spin $(k_1 + k_2 \neq 0, s + s' \neq 0)$. The model numerical calculations have shown that dependence of SC gap (energy of coupled states) on temperature is nonexponential and depends on the momenta of electrons in pair.

1. Introduction

The theoretical study of superconductivity (SC) is based as a rule on the physical $idea^{1-4}$ about pairing of electrons with the opposite spins (s+s'=0) and momenta (k+k'=0). Using this postulate the possibility of the appearance of the coupled states in the electron (electron-phonon) system was investigated. Those coupled states are responsible for superconductivity in the mentioned system.

It should be noted that coupled states can arise in the system of other type too. For example similar states occur in the phonons system of crystals [see Refs. 5, 6 and references therein] if anharmonic constant of forth order, responsible for phonon-phonon interaction is negative (V < 0). For this case pairs of coupled phonons may appear with full momentum K = k + k' equal to the momentum of incident on crystal photon. This type of excitation was observed in many spectroscopical experiments.^{5,6}

Absolutely similar situation must be realized for the electron system, if the interaction between identically charged particles due to some special mechanism became negative, i.e. electrons will attract each other.

In this communication a new approach to the SC problem is proposed. Namely we want to obtain the conditions when the coupled states in the electron system can appear, taking into account only interactions between quasiparticles of electron-phonon (e-ph) system. Our calculations have shown that hypothesis of electron pairing postulated in BCS theory is only approximate. That is the case of both momenta $(k+k'\neq 0)$ and so spins $(s+s'\neq 0)$. Our method of calculation is independent of whether the traditional SC or High T_c superconductivity (HTSC) crystals are investigated.

2. Short Theory

2.1. Density of current

In this paper we present the general idea of our approach. The operator which describes the density of current in the system of electrons can be written as follows

$$j'(x) = -c\frac{\delta H'}{\delta A(x)} = j(x) - \frac{e^2}{mc} A(x) \Psi^+(x) \Psi(x) ,$$

$$j(x) = -i\frac{e}{2m} \left\{ \Psi^+(x) (\nabla \Psi(x)) - (\nabla \Psi^+(x)) \Psi(x) \right\} ,$$
(1)

where operators $\Psi(x)$ are given by the following expression

$$\Psi(x) = \sum_{k,\sigma,\nu} a_{k,\sigma}^{\nu} \varphi_{k,\sigma}^{\nu}(x), \qquad (2)$$

and satisfy to Fermi anti-commutation relations

$$[\Psi(x,t), \Psi^{+}(x',t)]_{+} = \delta(x - x'), \qquad [a_{k,\sigma}^{\nu}, a_{k'\sigma'}^{+\nu'}]_{+} = \delta_{k,k'} \, \delta_{\sigma,\sigma'} \, \delta_{\nu,\nu'}, \varphi_{k,\sigma}^{\nu}(x) = \varphi_{k}^{\nu}(x) \chi_{\sigma}(S),$$
(3)

where $\varphi_k^{\nu}(x)$ is the Bloch function of crystal and $\chi_{\sigma}(S)$ describes the spin of electron in energy zone.

$$\rho(x) = en(x) = e\Psi^{+}(x)\Psi(x). \tag{4}$$

Then the density of current according to (1) is equal⁷

$$\langle j'(x)\rangle - \langle j(x)\rangle_{0} - \frac{e}{mc} \langle \rho(x)\rangle_{0} A(x,t) + \int_{-\infty}^{\infty} dx' dt' \left\langle \left\langle \left(j(x) - \frac{e}{mc} \langle \rho(x)\rangle_{0} A(x,t) \frac{e}{mc} \rho(x) A(x,t) \right), \right. \\ \left. \times \left(-\frac{1}{c} j(x') A(x',t') + \rho(x') \varphi(x',t') \right) \right\rangle \right\rangle_{0},$$
 (5a)

$$\langle \rho(x) \rangle_0 = \langle en(x) \rangle_0 = e \langle \Psi^+(x) \Psi(x) \rangle_0.$$
 (5b)

Here the expression (5a) is written up to linear terms on the components of electromagnetic field A(x,t), $\varphi(x,t)$; $\langle\langle \rangle\rangle_0$ is the retarded Green function from corresponding operators⁷ and index "0" points out on the thermodynamical equilibrium state.

The first term in Eq. (5a) is traditionally considered to be equal zero; the third have also studied (see e.g. Ref. 7). In this paper we shall investigate in detail the second term in (5a) given by (5b) and show that consistent taking into account the electron–electron (e–e) and electron–phonon (e–ph) interaction without any additional postulates (such for example as postulates in BCS theory about pairing of two electrons with opposite moments and spins) gives rise to coupled states in electron system and as sequence to superconductivity. Moreover from our aproach it follows that BCS postulates are approximate and they correspond to very simple limit case.

According to commutation relations (2) the value $\langle n(x)\rangle_0$ can be expressed by Fourier components of Green function in the following manner

$$\langle n(x)\rangle_0 = \langle \Psi^+(x)\Psi(x)\rangle_0 = \lim_{x'\to x, t'\to t-0} \left\{ \delta(x-x') - i\langle -iT\Psi(x,t)\Psi^+(x',t')\rangle \right\}$$

$$= \frac{\sum_k 1}{V} - i\sum_k G(k,\omega). \tag{6}$$

Here $\langle -iT\Psi(x,t)\Psi^+(x',t')\rangle$ is the Green function and $G(k,\omega)$ is its Fourier components. Our aim is to obtain this Fourier components with taking into account of e-e and e-ph interactions in crystals. So we find all spectral features mentioned system properties of which are determined by the following Hamiltonian.

2.2. Hamiltonian of crystal

The Hamiltonian describing the system of interacting electrons and phonons of crystal is written in the following form (further details will be shown elsewhere)

$$H = \sum_{k,\sigma,\nu} \varepsilon_k^{\nu} a_{k\sigma}^{+\nu} a_{k\sigma}^{\nu} + \frac{1}{N} \sum_{q,k_1,\sigma_1,\nu_1,k_2,\sigma_2,\nu_2} V_q a_{k_1\sigma_1}^{+\nu_1} a_{k_2\sigma_2}^{+\nu_2} a_{k_2+q,\sigma_2}^{\nu_2} a_{k_1-q,\sigma_1}^{\nu_1}$$

$$+ \frac{1}{\sqrt{N}} \sum_{k,\sigma,\nu,q,s} \chi_q^s a_{k,\sigma}^{+\nu} a_{k-q,\sigma}^{\nu} Q_q^s + \sum_{q,s} \Omega_{qs} h_q^{+s} h_q^s$$

$$(7)$$

where $V_q = V_q^*$, $X_q^{*s} = X_q^s$ are the Fourier components of Coulomb interaction of electron and their coupling constant with the lattice phonons respectively. We can see that both contants are independent on spin index of electron.

Lets us introduce a new operators for electron and phonon systems by rules

$$a_k^{\nu} = e^S A_{k,\nu} e^{-S}, \qquad b_q^s = e^S \beta_{s,q} e^{-S}$$
 (8)

where S is the anti-Hermitian operator $(S^+ = -S)$.

In our following unitary transformation it will be more convenient to rewrite the term H_{e-ph} in the next form

$$H_{\text{e-ph}} = \sum_{k,q,s,\nu,\mu} \chi_{s,q}^{\nu,\mu} a_k^{+\nu} a_{k-q}^{\mu} Q_q^s \,, \qquad (\chi_{s,q}^{\nu,\mu} = \chi_q^s \delta_{\nu,\mu}) \,, \tag{9}$$

where we united two indexes and so $\nu = (\nu, \sigma)$, $\mu = (\mu, \sigma')$ are the complex indices which characterize the number of crystal electron zone and the spin of electron.

The operator S we take as follows

$$S = \frac{1}{\sqrt{N}} \sum_{k,q,\nu,\mu} A_{k,\nu}^{+} A_{k-q,\mu} \sigma_{q}^{\nu,\mu} , \qquad \sigma_{q}^{\nu,\mu} = \sum_{s} \frac{1}{\Omega_{s,q}} \left(\chi_{s,-q}^{*\nu,\mu} \beta_{s,-q}^{+} - \chi_{s,q}^{\nu,\mu} \beta_{s,q} \right) . \tag{10}$$

In accordance to general rules of operator transformations (see e.g. Ref. 8 Appendix 5) the new reconstructed full Hamiltonian of e-ph system can be read as follows

$$H = \sum_{k,\nu} \left(\varepsilon_k^{\nu} - \frac{1}{N} \sum_{s,q} \frac{|\chi_q^s|^2}{\Omega_{s,q}} \right) A_{k,\nu}^+ A_{k,\nu} + \frac{1}{2N} \sum_{q,k,\nu,k'\nu'} \left(V_q - 2 \sum_s \frac{|\chi_q^s|^2}{\Omega_{s,q}} \right) A_{k,\nu}^+ A_{k',\nu'}^+ A_{k'+q,\nu'} A_{k-q,\nu} + \sum_{s,q} \Omega_{s,q} \beta_{s,q}^+ \beta_{s,q}^+ + \text{(higher-order term)}.$$
(11)

It is seen that fulfilled unitary transformation give rise to two principal effects: renormalization of electron energy (first term) and renormalization of Coulomb electron—electron interaction. In the later case if crystal contain many atoms in the unit cell effective Coulomb potential can became negative and significant in value.

2.3. Two-particle green function

The equation for Green function (6) with new operators is as follows:

$$i\frac{\partial}{\partial t}\langle -iTA_{k,\nu}(t)A_{k,\nu}^{+}(t')\rangle = \delta(t-t') + \left\langle -iT\left(i\frac{\partial}{\partial t}A_{k,\nu}(t)\right)A_{k,\nu}^{+}(t')\right\rangle, \tag{12}$$

where

$$i\frac{\partial A_{k,\nu}}{\partial t} = [A_{k,\nu}, H] = \tilde{\varepsilon}_k^{\nu} A_{k,\nu} + \frac{1}{N} \sum_{q,k_2,\nu_2} \tilde{V}_q A_{k_2,\nu_2}^+ A_{k_2+q,\nu_2} A_{k-q,\nu}$$

$$= \tilde{\varepsilon}_{k,\nu} A_{k,\nu} - \frac{1}{N} \sum_{q,k_2,\nu_2} \tilde{V}_q A_{k+q,\nu} A_{k_2-q,\nu_2} A_{k_2,\nu_2}^+, \tag{13}$$

$$\tilde{\varepsilon}_{k}^{\nu} = \varepsilon_{k}^{\nu} - \frac{1}{N} \sum_{s,q} \frac{|\chi_{q}^{s}|^{2}}{\Omega_{s,q}}, \qquad \tilde{\varepsilon}_{k,\nu} = \tilde{\varepsilon}_{k}^{\nu} + \sum_{\nu_{2}} \left(\tilde{V}_{q=0} - n \frac{1}{N} \sum_{q} \tilde{V}_{q} \right),$$

$$\tilde{V}_{q} = V_{q} - 2 \sum_{s} \frac{|\chi_{q}^{s}|^{2}}{\Omega_{s,q}}.$$

$$(14)$$

The last line in Eq. (13) was obtained by using the commutation rules (3) for Fermi operators A_{kj} .

Therefore the equation for Green function (12) is transformed after inserting in it (13) to the following

$$i\frac{\partial}{\partial t}\langle -iTA_{k,\nu}(t)A_{k,\nu}^{+}(t')\rangle = \delta(t-t') + \tilde{\varepsilon}_{k,\nu}\langle -iTA_{k,\nu}(t)A_{k,\nu}^{+}(t')\rangle - \frac{1}{N} \sum_{q,k_{2},k_{2}} \tilde{V}_{q}\langle -iTA_{k+q,\nu}(t)A_{k_{2}-q,\nu_{2}}(t)A_{k_{2},\nu_{2}}^{+}(t-0)A_{k,\nu}^{+}(t')\rangle.$$
 (15)

To calculate the density of electron states we have to consider the case of striving for $t' \to t - 0$ in accordance with (6). For this situation the last (four operator) Green function in the Eq. (15) can be written as follows

$$G_{2}\begin{pmatrix}k_{2}, \nu_{2}; k, \nu\\ k+q, \nu; k_{2}-q, \nu_{2} \end{pmatrix} t - t'$$

$$= \langle -iTA_{k+q,\nu}(t)A_{k_{2}-q,\nu_{2}}(t)A_{k_{2},\nu_{2}}^{+}(t')A_{k,\nu}^{+}(t')\rangle \qquad (t' \to t - 0). \quad (16)$$

Our aim is now to find the function (16) and using the relation between Fourier components of this function and one-particle electron function following from Eq. (15) to calculate the density of the electron states.

In this paper we will study only the spectral properties of two-particle Green function (16). Following to the approach used for phonon system⁵ which is based on the method of Bogolubov-Tyablikov^{9,10} (the details of calculation will be given in special communications) we have derived the equation for two-particle electron Green function. The spectral features of electron system in the mentioned region of energy are described by Fourier component of this function and for the simplest case of one-electron zone crossing the Fermi energy level can be given by the following expression

$$\Phi(k_1, k_2, \omega) = \frac{f(k_1, k_2, \omega) \sum_{\sigma, \sigma'} \varphi(\sigma, \sigma')}{1 - V \sum_{q} (1 - n_{k_1 + q} - n_{k_2 - q}) / \omega - \varepsilon_{k_1 + q} - \varepsilon_{k_2 - q}},$$

$$V = \tilde{V}_q = V_q - 2 \sum_{s} \frac{|\chi_{s,q}|^2}{\Omega_{s,q}} \approx \text{const}.$$
(17)

 n_k is the filling number of electrons; V is the effective Fourier component of electronelectron (e–e) interaction. If the constant of e–e interaction renormalized by e–ph interaction becomes negative (V < 0) the arising of coupled states in the electron system is possible.

It is seen from Eq. (17) that for $k_1 = k_2 = 0$ or $k_2 = -k_1$ the denominator is reduced to well known expression describing the SC gap of crystal, but for other arbitrary meanings of momenta k_1 and k_2 the situation becomes significantly different.

Besides the numerator of Eq. (17) gives rise to new possibilities. Here $f(k_1, k_2, \omega)$ is the some function depending on the frequency (ω) and momenta k_1 and k_2 of pair of electron; the second function $\varphi(\sigma, \sigma')$ (which is written for one band crossing the Fermi level) is expressed by number of delta-functions $\delta_{\sigma,\sigma'}$

$$\varphi(\sigma, \sigma') = \delta_{\sigma\sigma} \delta_{\sigma'\sigma'} - \delta_{\sigma\sigma'} \delta_{\sigma\sigma'} , \qquad (18)$$

where σ, σ' are spines of first (σ) and second (σ') electrons respectively. We can see that two cases are possible:

- (a) if spins of electrons are parallel to each other $(\sigma = \sigma')$ then $\varphi(\sigma, \sigma') = 0$;
- (b) if spins of pair of electrons are antiparallel to each other $(\sigma \neq \sigma')$ the second term in the right hand of (18) (which is proportional to $\delta_{\sigma,\sigma'}$ disappear and therefore $\varphi(\sigma,\sigma')\neq 0$). It means that only states with the oppositely directed spins, namely singlet states $(\sigma+\sigma'=0)$ in this simplest case, can contribute into coupled states of electron system. For more complex structure of crystal zones in particular for layer Bi-based type of crystal for example the function $\varphi(\sigma,\sigma')$ in (17) will be more complicated. This situation will be studied elsewhere.

2.4. Coupled states and gap

The energy position of coupled states are given by the denominator of (17).

Let us study the situation near the extremum (minimum or maximum) of the electron zone. Namely we will suppose $k_1 = k_2 = k_0 + k$ and $\varepsilon_{k_0,\mu} = \varepsilon_{\mu}$ corresponds to extremum of zone. Then expanding the energy by momentum $k \pm q$ in series up to terms of second order and supposing that energy extremum is located near the region of Fermi level energy we can obtain

$$\varepsilon_{k \pm q, \mu} = \varepsilon_{\mu} + \frac{(k \pm q)^2}{2m_{\mu}} = \varepsilon_{\mathbf{f}} + \Delta_{\mu} + \frac{(k \pm q)^2}{2m_{\mu}}, \qquad (\mu = 1, 2,), \tag{19}$$

where m_{μ} is the effective mass of electron in the μ th energy zone of crystal; Δ_{μ} is a parameter which points out the position of the μ -zone extremum relative to Fermi level. It may be negative and positive and the effective mass the same. This dispersion law for example describes two type of electron zones crossing the Fermi level in the Bi-based crystals. Taking in mind such type of crystal as an example and using the Eq. (19) we will study the conditions of arising the coupled states for the several cases and compare the analytical results with the numerical calculations.

Here we may analyze it in the traditional manner but taking into account also the influence of states with the $k \neq 0$. The sum in the denominator (17) is reduced to the following expression (we consider only one zone)

$$K(k, k', \omega) = \frac{1}{N} \sum_{q} \frac{1 - n_{k+q} - n_{k-q}}{\omega - \varepsilon_{k+q} - \varepsilon_{k-q}}$$

$$= 2N(\varepsilon_f) \int_{0}^{\Delta} \frac{d\varepsilon}{\omega - 2(\varepsilon_f + \Delta_1) - 2E - 2\varepsilon} = N(\varepsilon_f) \left(-\ln\left|1 - \frac{\Delta}{a}\right| \right), \tag{20}$$

where such assignment were used: $\varepsilon = q^2/2m$; $E = k^2/2m$; $a = \omega - 2(\varepsilon_f + \Delta_1) - 2E$; $m_1^* = m_1/m$ and m are the reduced effective mass of electron in the crystal energy zone and mass of free electron respectively.

$$N(\varepsilon_f) = \sqrt{2\pi} m_1^* \sqrt{m_1^* \varepsilon} (1 - n_{k+q} - n_{k-q}) \Big|_{\varepsilon_f},$$

$$n_k = \left[\exp\left(\frac{\varepsilon_k - \varepsilon_f}{T}\right) + 1 \right]^{-1}.$$
(21)

The equation for coupled states in the electron system has the following form

$$1 - VN(\varepsilon_f) \left[-\ln \left| 1 - \frac{\Delta}{a} \right| \right] = 0.$$
 (22)

We can see from Eq. (20) that influence of momenta $(k \neq 0, \text{ i.e. } E)$ and parameter Δ_1 is identical (but the later may be positive and negative) therefore their influence can be simply analyzed by graphically using the Eq. (22). We will consider two cases presented in Fig. 1.

- (a) The case k=0 (E=0) is pictured by crossing of curves 1,1' (they correspond to the function $-N(\varepsilon_f) \ln |1-\Delta/a|$) with the horizontal line 4 (it corresponds to value 1/V, V<0). The graphical solution gives points A and A'.
- (b) The cases $k = k_1 \neq 0$ and $k = k_2 \neq 0$ correspond to the crossing of curves 2, 2' and 3, 3' with the horizontal line 4 in points B, B' and C, C' accordingly.

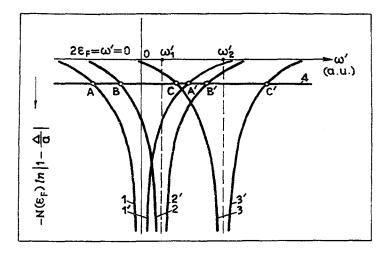


Fig. 1. The graphical solution of Eq. (22): curves 1, 1'-3, 3' describe the function $(-N(\varepsilon_f) \ln |1-\Delta/a|)$; curve 4 describes the constant (-1/|V|); points A,B correspond to the coupled states of electron system.

We can see in Fig. 1 that only points A and B are located below $2\varepsilon_f$ energy level. Namely these points corresponds to coupled states. For other states which

correspond to the points A', B', C and C' in Fig. 1 coupling is impossible. Therefore we may conclude that there are group states with nonzero momenta $(k \neq 0)$ which are important due to their significant influence on the SC state of electron system.

The analytical expression for energy position of coupled states ($\omega' = \omega - 2\varepsilon_f < 0$) taking into account the condition $k \neq 0$ is given by

$$\omega' = \omega - 2\varepsilon_{\rm f} = 2(E + \Delta_1) \mp 2\Delta / \left[\exp\left(\frac{1}{|V|N(\varepsilon_{\rm f})}\right) \mp 1 \right].$$
 (23)

It is clearly seen (in agreement with the above draphical analysis) that only for some region of impulses $k < k_{\rm max}$ (here $E_{\rm max} = k_{\rm max}^2/2m$) the right hand part of (23) can be negative, i.e. the coupled states can emerge. This region may be essential and this depends on the value of |V| effective constant of e-e interaction (and also on parameter Δ_1). The higher these values the greater number of states with the $k \neq 0$ give the contribution to the coupled states of electron system.

2.5. Numerical calculations

The analytical analysis indeed not taking into consideration many features which are contained in the denominator of Eq. (17) because of the influence of momenta and temperature by the electron filling number were excluded. It should be noted that for some type of crystals (Bi-based for example Ref. 11) the minimum of electron zone can be located much lower (0.7–0.8 eV for $Ba_2Sr_2CaCu_2O_8$) of the Fermi level energy. Therefore it is interesting to consider namely such situation because this crystal has a high T_c temperature.

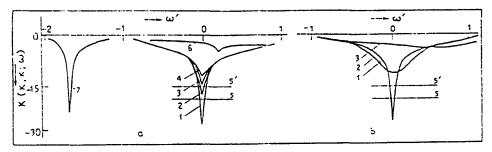


Fig. 2. The calculations of the first sum in Eq. (20) and dependences of coupled states on temperature and momentum: (a) $\Delta_1=-1,\ E=0$ curve 1, T=2 K; curve 2, T=10 K; curve 3, T=50 K; curve 4, T=100 K; curve 6, $\Delta_1=+0.1,\ E=0$ (the curves for all temperatures considered are coincide); curve 7, corresponds to analytical consideration (see text). (b) $\Delta_1=-1,\ T=2$ K: curve 1, E=0; curve 2, E=0.001; curve 3, E=0.05.

The numerical calculations including these factors give rise to the following results.

The influence of parameter Δ_1 (when k=0) it is seen in Fig. 2a (for calculations we use parameters $\Delta_1=(\Delta_1/M)=1, +0.1; M=1$ eV; $m_1^*=1$) where some curves describing the first sum of the Eq. (20) with taking into account (19)

at different temperatures are presented. It is seen in Fig. 2a that increasing of the temperature (in region 1-100 K) gives rise to significant decreasing of depth of minima. The crossing these curves with the horizontal lines 5 (V = V/M = -0.05) and 5' (V = -0.06) corresponds to meanings of energy at which denominator of Eq. (17) is zero. It follows from calculations that for temperature T > 60~K(for given values parameters of crystal) the crossing is absent and coupled states (responsible for SC) are impossible. At very weak coupling constant (V < 0.04) the coupled states disappear at T > 10 K. For stronger values (V = -0.05 - 0.07) the critical temperature T_c increases to 60–100 K.

The curves 1-4 in Fig. 2a are obtained at $\Delta_1 = -1$. If this value comes to zero and then becomes positive the depth of minimum studied curves becomes much smaller and coupled states disappear even at low temperature. That illustrates curve 6 in Fig. 2a. Therefore we can conclude that location of the electron energy zone minimum relatively to Fermi level is very important and coupled states as rule cannot arise if parameter $\Delta_1 \geq 0$.

The curves in Fig. 2b demonstrate the influence of momentum k (or E) on the arised coupled states. It is seen in Fig. 2b that curves 1-3 are different in form and its maxima are shifted to high frequency region. The depth of minimum is decreasing for great momentum k. The results of numerical calculations show that there is some region of momenta $k < k_{\text{max}}$ which gives the contribution into coupled states. In particular that very important for complex structure of crystal having some electron zones crossing the Fermi level energy (these aspects will be analyzed elsewhere).

If we exclude the temperature factor (the numerator in Eq. (20) is believed constant) the curve 1 in Fig. 2a is transformed into curve 7 in Fig. 2a which are in correspondence with the results of Fig. 1 and the traditional analytical calculations. 1-3 Therefore we can conclude that only taking into account all factors (temperature, momenta, effective mass etc.) give rise to correct results in calculation of SC temperature (T_c) in crystal. As a result the energy of the coupled states (SC gap) is not described by the exponential dependence similar to Eq. (23). It should be noted also that calculated SC gap is dependent on momentum k according to Eqs. (17) and (20) must be averaged on all momenta of electrons as it made in our approach. It is obvious therefore that mentioned above factors can result in to complicated dependence of SC state of crystal on temperature not described in the framewok of BCS theory.

More detailed analysis of other features for complex crystals, in particular, states with the nonzero spins $(s + s' \neq 0)$ will be discussed elsewhere.

3. Conclusion

The new theoretical approach is proposed for the description the coupled states in the many electron system. It is shown that postulates on electron pairing which are used in BCS model are only approximate. New more rigorous rules are derived. It is shown that influence of electron pairs full momentum of which is nonzero $(k+k' \neq 0)$ may be significant in the superconductivity (and also HTSC) of electron system.

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