d-ELECTRONS AND SUPERCONDUCTIVITY OF TRANSITION METAL OXIDES

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Some problems of the normal state of high temperature cuprate superconductors (HTS) with *d*-wave pairing are discussed. It is shown that: 1) at pseudogap temperature T^* it occurs the dimensional crossover of the normal incoherent state to quasi 2D system of $Cu\ O_2$ layers; 2) «naked holes» are bad quasiparticles for quasi 2D system with Jahn-Teller and mixed-valent *Cu* ions, with strong electron correlations and fluctuations, and with inherent strong *p-d* hybridization of copper and oxygen orbitals. Under these circumstances the holes are reduced to two types of the magneto-elastic polarons: the ferromagnetic bound polarons, which lead to forming of the stripes state, and antiferromagnetic Jahn-Teller-Zhang-Rice polarons which can form pairs and lead to the superconductivity.

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INTRODUCTION

A few theoretists are succeeded in the prediction of new phenomenons. But academician A.I. Akhiezer was namely such happy scientist. In solid status physics a lot of his predictions were realized: magnetic-acoustic resonance (together with V.G. Baryakhtar and S.V. Peletminskij, 1958); known among experimentalists “Akhiezer attenuation”, etc. But here it will talk about long and successful life of his works with I.Ya. Pomeranchyk[1], and with I.A. Akhiezer[2] where *p*-wave superconductivity was predicted. It was very courageous prediction because only in 1960 the paper of Mathias and Suhl was published where at first it was shown that superconductivity and ferromagnetism are coexisting in doped Ru. They wrote: “In the range of $T<T_c$ superconducting regions are coincident with domain walls, and form an intricate honey-comb or “sponge”…”.

In first paper [1] the interaction of *s*-electrons with *d*-electrons by means of ferrromagnetic spin waves exchange was studied, and it was shown that it leads to the attraction between *s*-electrons, and to the superconductivity. A.I. Akhiezer and I.Ya. Pomeranchyk wrote:“The attraction between *s*-electrons has place in the triplet state. The dimensional part of the wave function is antisymmetric (*p*-state).”

Thirty six years were over, and only in 1995 T.M. Rice and M.J. Sigrist shown that superconductor $Sr_2Ru\ O_{4.1}$ has the triplet gap with *p*-symmetry of order parameter: *p*-wave pairing of *s*-electrons occurs by means of spin waves exchange with *d*-electrons [3]. Thus, at once three predictions of A.I. Akhiezer and I.Ya. Pomeranchyk were putted into practice:

i) for *d*- and *s*-electrons exchange by spin waves of ferrromagnetic type causes the attraction of *s*-electrons;
ii) the attraction leads to superconductivity;
iii) the order parameter has *p*-symmetry.

Crucial role of *d*-electrons in the unusual properties of transition metal compounds at least fifty years is intensively discussed in condensed state physics. The interest to them is caused by two important problems with studying of the nature of colossal magneto-resistance materials, such as manganites; and nature of high temperature superconductivity in general, and essentially for cuprates HTS’s with *d*-wave pairing.

Nearly 15 years high temperature superconductivity still remains not being understood theoretically. And yet new questions are now addressing to the normal state of HTS’s:

i) what is the nature of the “pseudogap” state at $T^*>>T_c$?
ii) what is the “stripe states”?

This paper is an attempt for underdoped HTS to discuss these problems at the supposition that at temperature T^* the dimensional crossover of the normal state from 3D incoherent state to quasi 2D system of $Cu\ O_2$ layers occurs. For such systems with Jahn-Teller and mixed-valent Cu^{+2} and Cu^{+3} ions strong electron correlations and fluctuations, and inherent strong *p-d* hybridization of copper and oxygen orbitals are making bad quasiparticles out of a “naked holes” and lead to their reducing to magneto-elastic polarons.

DUAL CHARACTER OF *p-d* HYBRIDIZATION IN PEROVSKITES OF TRANSITION METALS

First of all it is need to understand why the same *d*-electrons in $Sr_2Ru\ O_{4.1}$ lead to *p*-wave pairing, and in cuprate HTS’s, for example in La_2CuO_{4+x} , they lead to *d*-wave pairing. The cause of this difference is lying in the crucial role of *p-d* hybridization for these compounds. It is known that oxides of transition metals (TM) have inherent strong *p-d* hybridization of TM and oxygen orbitals with dual character of results: π-bonds with $(d_{xy} - p)$ hybridization lead to direct ferromagnetic (FM) exchange *(I<0),* and *p*-wave superconductivity, and σ -bonds with $(d_{x^2-y^2} - p)$ hybridization lead to the indirect antiferromagnetic (AFM) exchange *(J>0),* and *d*-wave superconductivity. Symmetry of the order parameter depends on the bonds character of the states near Fermi energy E_F: for cuprates they have $(d_{x^2-y^2} - p_\sigma)$ character, and $J>>I$, that leads to *d*-wave superconductivity; but for ruthenates they have $(d_{xy} - p)$ character, and *I>>J,* that leads to *p*-wave superconductivity. In the simple *t-J-I* model of *p-d* hybridization [4] with AFM and FM interactions it was shown that the temperature of the superconducting transition with *p*-wave pairing, $T_c(p)$, is more less than the temperature $T_c(d)$ of the transition with *d*-wave pairing

$$T_c(p) << T_c(d)$$

at the equal interaction constants.

The main problem in the normal state of HTS is its incoherent behavior, which point out that «naked holes» are bad quasiparticles because they are strong interacting with localized spins of copper and moving oxygen ions. At first Landau [5] point out that even in ideal crystal strong interactions between quasiparticles and moving ions lead to new type of quasiparticles, which later were named polarons. Magnetic polarons at first were introduced by Nagaev in 1967 (see refs. in review [6]).

It is known that for perovskite copper oxides there are two type polarons: inherent strong *p-d* hybridization of ions orbitals leads to AFM Zhang-Rice polarons (ZRP's) [7], and FM polarons (FMP) [6]. Intermediate size FMP's and phase separation in the copper oxides were investigated in ref.[8]. Existence of localized polalaronic states depends on the dimensionality of system and on the value of the amplitude of the *p-d* hybridization.

The charge transfer along c-axis for underdoped HTS has incoherent character and it is the result of thermal fluctuations at the temperatures

$$k_B T > t_c^2(T)/t_{ab}\,. \tag{1}$$

Here t_c and t_{ab} are the strength of interlayer and intralayer couplings of the charges, k_B is Boltzman constant. At the temperature decreasing thermal fluctuations limit out the interlayer tunneling, and at

$$k_B T \approx t_c^2(T)/t_{ab} \tag{2}$$

this leads to the dimensional crossover when at the temperature

$$T^* = t_c^2(T^*)/k_B t_{ab}$$

the charge in *Cu* O_2 plane becomes "two-dimensional". At $T<T^*$ the change of the metallic character of the resistivity along c-axis on the semiconductive one oc curs. The conclusion about the dimensional crossover to the system of quasi 2D *Cu* O_2 planes is very important because for 2D systems any localized states (including polaronic states) exist at the any value of the interaction constants. At once one can see that the value of the temperature of the dimensional crossover T^* depends on doping concentration for all cuprates: the dopants take up the positions between the *Cu* O_2 planes, and strong affect the charge transfer along c-axis.

CHARGE HAMILTONIAN IN *CuO₂* PLANE

We consider a Hamiltonian describing a charge in single CuO_2 layer

$$H_{ef} = H_{DE} + H_{JT}\,, \tag{3}$$

where

$$\begin{aligned} H_{DE} &= \frac{1}{2}\sum_{\rightarrow} J_{n,n+\rho}\vec{S}_n\vec{S}_{n+\rho} - \\ &- J_H\sum S_n a^+{}_{n,\lambda,\sigma}\hat{\sigma} a_{n,\lambda,\sigma} - \sum t^{\lambda\lambda'}_{n,n+\rho} a^+_{n,\lambda\sigma} a_{n+\rho,\lambda'\sigma} \end{aligned} \tag{4}$$

Here H_{DE} is the phenomenological Kondo Hamiltonian with the super exchange interactions $J_{n,n+\rho}$ of the neighboring *Cu* ions spins S_n through the oxygen ions p_σ bonds, which have AFM character, $J_{n,n+\rho}$ >0, and the superexchange interactions through the p_π bonds with the FM character [9], $J_{n,n+\rho} \equiv I_{n,n+\rho} < 0$. The former AFM interaction leads to the effective integral of the charge transfer $t^{\lambda\lambda'}_{n,n+\rho} = t_{AFM}$. Here t_{AFM} is the hybridization amplitude, which is proportional to the overlap of the orbitals of the wave functions ($d_{x^2-y^2}, p_\sigma$). The FM interaction leads to the charge transfer $t^{\lambda\lambda'}_{n,n+\rho} = t_{FM}$, where t_{FM} is proportional to the overlap of the orbitals (d_{xy}, p_π) of the wave functions.

In (4) second term $\boldsymbol{J_H}$ is the Hund exchange, $a^+_{n\lambda\sigma}, a_{n\lambda\sigma}$ are the hole operators, corresponding to the hole at the atom *n* with the spin σ in the state λ , and third term is the tunnel charge transfer between *Cu* ions.

Second component in (3) is the phenomenological Hamiltonian of the charge interaction with Jahn-Teller distortion of oxygen ions:

$$\begin{aligned} H_{JT} &= g_{JT}\sum_{n\lambda\lambda'\sigma}[a^+_{n\lambda\sigma}Q_n^{\lambda\lambda'}(j)a_{n\lambda'\sigma} + \\ &+ \frac{k_{JT}}{2}Q_n^2(j) + \frac{M_{JT}}{2}(dQ_n/dt)^2] \end{aligned} \tag{5}$$

Here g_{JT} is constant of the elastic Jahn-Teller (JT) interaction, $Q_n(j)$ is operator of *j* normal JT modes, k_{JT} is their elastic constant and M_{JT} is the effective mass.

The canonical Holstein-Lang-Firsov transformation [10] let us in (5) to get rid of first term, and leads to the new stationary states which are magneto-elastic Jahn-Teller polarons with renormalizated integral of the charge transfer

$$t \Rightarrow t_{JT} = t\exp[-\frac{g^2_{JT}}{\Omega^2_{JT}}(1+2n_{JT}] \tag{6}$$

Here n_{JT} is mean phonon number with frequency

$$\Omega_{JT} = \sqrt{k_{JT}/M_{JT}}\,.$$

From (6) one can see that the new quasiparticles are realy depending on the magnetic properties (through the values of the charge transfer), and on the phonons frequencies Ω_{JT}, and on the JT interactions constant.

JAHN-TELLER RENORMALIZATION OF ZHANG-RICE POLARONS

The superexchange interaction $J_{n,n+\rho}$ of spins S_n of the neighboring *Cu* ions with oxygen ions through p_σ bonds $(d_{x^2-y^2} - p_\sigma)$ has AFM character and is equal [7]

$$J^{AFM}_{n,n+\rho} = \frac{4\tilde{t}^{4}_{AFM}}{\varepsilon^{2}_{p}}(1/U + 1/2\varepsilon_p) > 0 \qquad (7)$$

Here $\tilde{t}_{AFM}$ is effective integral of the charge transfer t_{AFM} with taking into account the renormalization (6); $\varepsilon_p > 0$ is the atomic energy of oxygen hole; and *U* is the Coulomb repulsion at *Cu* site [11].

As it was shown by F.C. Zhang and T.M. Rice [7] *p-d* hybridization strongly binds a hole on each square of O^{-2} ions to the central Cu^{+2} ion to form a local singlet, Zhang-Rice polaron Taking into account the renormalization (7) of the AFM interaction $J^{AFM}_{n,n+\rho}$ let us below to name this local singlet as Jahn-Teller Zhang-Rice polaron (JT ZR polaron). This JT ZR polaron is singlet state in which *p*-wave *O* holes is combined with *d*-wave *Cu* hole, and the large binding energy in the singlet state is due to the phase coherence. This polaron has the energy

$\approx 15{,}68\,(\tilde{t}_{AFM})^2(\varepsilon_p)^{-1}$,

and moves through the lattice with effective nearest-neighbor hopping

$t_{ef} \approx 1.5(\tilde{t}_{AFM})^2(\varepsilon_p)^{-1}$.

The depth of JT ZR polaron level is exceeded *d-d* exchange energy $E_{dd} = 4JS^2$. JT ZR polaron has the important advantage over «naked hole», because its state is the phase coherent state; therefore JT ZR polarons are really good quasiparticles with large value of coupling with phonons. This large coupling is obtained at small holes concentration $n<n_c$ where $n_c \sim 10^{20}\ \mathrm{cm}^{-1}$. It is known that at this condition plasma edge lies below the highest optical phonons and they are unshielded**.**

JAHN-TELLER RENORMALIZATION OF BOUND FERRONS

The studying of FM self-trapped states of a charge carriers in the doped AFM crystal was began by E.M. Nagaev which in 1968 proposed the models of free FMP and bound ferron (see refs. in [6]). For free ferron in layered AFM crystal strong magnetic anisotropy along c-axis leads to the absence of well defined FM region with a finite size inside of which the electron is localized. In the frame of I.M. Lifshits theory for disordered 2D system the discrete level exists at any values of *t* and *I* [12]. But it is too shallow to compensate the loss in the *d-d* –exchange energy, because the quantity of the ratio

$$|I/J| \approx J_H/U \leq 0.2$$

is small, and leads to the depth of the level which is insufficient for this compensation. Therefore for doped layered anisotropic antiferromagnets the bound ferrons consideration is more accurate [6]. The bound ferron is corresponding to a renormalized hole which is localized at an impurity neighborhood. Creation of a magnetized region around the impurity additionally diminishes the energy of the system. For cuprate HTS's ions Cu^{+n} ions with $n \neq 2$ can serve as the localization centers.

In Ref. [6] it was shown that electron wave function $\Psi(r)$ in the magnetized region should be determined from the same equation as for the electron in a hydrogen atom with an additional requirement

$\Psi(R)=0$

at *r=R,* where *R* is the radius of the FM microregion:

$$(-\Delta/2m - e^2/r\varepsilon - U - E_s)\Psi(r) = 0. \qquad (8)$$

Here E_s is the energy of the bound state, and

$m=1/\tilde{t}_{FM}a^2$

is the electron effective mass, ε is the dielectric constant, *a* is *Cu-O* distance in $Cu\,O_2$ plane, and $U=2\tilde{t}_{FM}$ is the spherical potential well. $\tilde{t}_{FM}$ is effective integral of the charge transfer t_{FM} with taking into account the renormalization (6).

The ground state wave function $\Psi(r)$ of Eq. (8) is given in Ref. [13], and expresses through the confluent hyper-geometric Kummer function

$\Phi(1-n, 2, 2r/na_B)$,

where *n* is the argument of this function, and a_B is the Bohr radius[14]. Analysis of these functions shown, that the minimal ferron energy is lower than the energy of the bound *s* electron, and the size of bound ferron is equal

$$R \cong 2\frac{\varepsilon\tilde{t}_{FM}a^2}{e^2}. \qquad (9)$$

With taking into account value $\varepsilon = 9$ of the dielectric constant for CuO_2, we can estimate the value

$R\sim 2\varepsilon a_B \geq 10$ Å.

If the size of the bound ferron is compared with the mean distance between the neighboring localization centers, Cu^{+n} ions, $R\sim ax^{-1/3}$, (*x* is the doping concentration), the chain of the bound ferrons leads to generation in CuO_2 planes of the net of the stripes along which can move only pairing polarons, because the depth of the JT ZR polaron level is sufficient to destroy the potential well which was created by the bound FM polarons. With using of the equation (9) we can estimate the critical value x_{cr} of the concentration at which the bound ferrons don't overlap

$$x \leq x_{cr} \sim (a/R)^3 \sim e^6/(2a\varepsilon\tilde{t}_{FM})^3, \qquad (10)$$

because their overlap at $x>x_{cr}$ leads to the destruction of the bound ferron states.

DISCUSSION

For J.G. Bednorz and K.A. Muller the idea that the Jahn-Teller polarons with strong electron-phonon coupling might be important for high temperature superconductivity was the original direction of their searches [15]. The existence of the attraction interaction between the polarons and their pairing with forming bipolarons for TM oxides, and ceramics $Ba\,Bi_x\,Pb_{1-x}\,O_3$

was firmly determined 20 years ago (see for example reviews [16,17]). It is important to note that for TM oxides the observation of the charge ordered state is bound up with bipolarons [17]. At first the conditions of the transition to supercoducting state of systems with the bipolarons at the exchange by optical phonons were discussed in Ref.[18], and for HTS's the polaronic pairing mechanism now are intensively discussed (see review [19]).

It is shown above that dual character of the *p-d* hybridization in perovskites of transition metals leads to two types of the polarons. One of them is the mobile AFM Jahn-Teller-Zhang-Rice polaron, and second is bound FM polaron (or Nagaev bound ferron[6]). Below it will be shown that both types of the polarons are very important for HTS's.

1. Stripes generation for underdoped HTS.

In this paper it was shown that inherent strong *p-d* hybridization without fail leads to the strong electron-phonon coupling which caused by moving oxygen ions. At $T<T^*$ charge becomes "two-dimensional", and the strong Jahn-Teller distortion, and the $(d_{xy}-p)$ hybridization lead to the direct FM exchange between orbitals of the *d*-electrons of the neighbor *Cu* ions and to the generation of the bound ferrons in $Cu\ O_2$ plane. For the underdoped HTS's with dopant concentration $x \leq x_{cr}$ this leads to the generation of the stripe structures in $Cu\ O_2$ plane.

For the generation of the stripes condition (10) is very important because the overlap of the bound ferrons leads to the decreasing of the bound level depth and to loss of the compensation of the *d-d* exchange energy, and to the destruction of the bound ferron states. As it is seen from (10) the critical value x_{cr} depends only from the parameters of $Cu\ O_2$ planes. This means that for all HTS cuprates the region of the stripe state on phase diagram is bound by the same value of the holes number on one *Cu* ion.

Another important condition for stable stripes state is the pairing state of JT ZR polarons. It was shown in ref.[3] that two holes are strong repulsing on the same square, because the energy of two separated holes is smaller than the energy of the two holes residing on the same square. This leads to the creation of two neiboring JT ZR polarons which can form stable intersite bipolaron if their coupling with phonons is sufficiently strong and can overcome the Coulomb repulsion [18,19]. The A.S. Aleksandrov [19] estimations for small polarons are confirmed by the numerical simulations of ionic perovskite lattices which established the existence of stable intersite bipolarons in doped cuprates [20]. Here it is shown that JT ZR polarons are small ones, and they form bipolarons which can freely move without the destruction of the chains of the bound ferrons.

2. JT ZR polarons pairing as a mechanism of the high temperature superconductivity.

The bipolarons forming in the $Cu\ O_2$ plane can lead to the two dimensional superconducting fluctuations with the value of the coherent length in the CuO_2 plane

$$\xi_{ab}(T) = \xi_{ab}(T_{BKT})(T/T_{BKT}-1)^{-1/2},$$

where T_{BKT} is the two dimensional superconductivity temperature Berezinskii-Kosrelitz-Thouless (BKT), and to the decreasing of the tunneling probability of the charge along c-axis

$$t_c(T) = \frac{\xi_c^2}{\xi_{ab}^2}(T/T_{BKT}-1), \qquad (11)$$

where ξ_c are the values of the coherent length along c axis at $T = T_{BKT}$, [21]. At sufficiently small $t_c(T)$ the Kats inequality [22]

$$T_c / E_F \geq t_c(T_c) \qquad (12)$$

determines the temperature of supeconducting transition which occurs as two dimensional one with small region of 3D superconducting fluctuations. Thus, from the eqs. (11) and (12) we can receive the estimation

$$T_{BKT} \leq T_c \leq \frac{\xi_c^2 E_F T_{BKT}}{\xi_c^2 E_F - \xi_{ab}^2 T_{BKT}} \qquad (13)$$

Thus, the attraction of mobile JT ZR polarons can leads to the high temperature superconductivity at the conditions that the frequencies of Jahn-Teller cooperative oxygen displacements, and the energy of electron-phonon coupling, and the Coulomb energy are the values of the same order [18]. This means that all these interactions together are important participants of the high temperature superconductivity. These circumstances make the searches of pairing mechanism for HTS's such prolonged and hard work.

3. About the experimental observations of the bipolarons for cuprates HTS's.

In Refs. [8, 22-23] this possibility was discussed, and it was shown that polaron states might be identificated from optical absorption data. At the studying optical absorption and reflectivity spectra of layered copper oxides $LaCuO_{4+x}$ over 2 eV the dominant contribution of polarons was found out [23]. Recently [24] at the studying the difference absorption spectra of the monocrystilline films $YBa_2Cu_3O_{6+x}$ (x =0.3) in AFM phase the narrow absorption band at region 2.0÷2.2 eV was observed at $T<200$K. This observation one can consider as the absorption at the generation of JT ZR polaron. In the metal phase of YBCO with x =0.85 the absorption spectra at the region 2.0÷2.4eV has the doublet structure with maxima at 2.145eV and 2.28 eV, that appears abruptly in the temperature region of the formation of the stripes structure at $T<T^*$. This observation one can consider as the absorption at the bipolaron dissociation in $Cu\ O_2$ planes on two JT ZR polarons. For this consideration we have next evidences [25]: i) the dispersion of each of the doublet components is equal of one for the narrow absorption band for the sample in AFM phase (x =0.3); ii) the additional component at $T<T^*$ appears from the high frequency side of latter that is a sign of the AFM nature of the polarons. At $T=T_c$ the additional increase of the each maximum amplitude occurs that evidences about the transition in 3D superconducting coherent state that leads to the increase of the bipolarons number. The amplitude of each maximum does not change at the

$T<T_c$ that evidences about the bipolarons existence in the superconducting state.

In conclusion, the author want to remark that it is a great honor for her to be a participant of A. Akhiezer Memorial Conference, and that it was a happy chance for her to work and to associate with this Eminent Person.

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