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CONDENSED MATTER

Stability of the Superconducting $d_{x^2-y^2}$ Phase in High-T_c Superconductors with Respect to the Intersite Coulomb Repulsion of Holes at Oxygen

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It has been shown that, because of the two-orbital character of the subsystem of holes located at oxygen sites and the spatial separation of this subsystem from that of spins at copper ions, the superconducting phase in high-T_c superconductors is stable with respect to the strong Coulomb repulsion of holes located at nearest-neighbor oxygen sites if the order parameter has the $d_{x^2-y^2}$ symmetry. This effect is due to the symmetry characteristics of the Coulomb potential, owing to which the equation determining the Cooper pairing in the $d_{x^2-y^2}$ channel does not include this potential.

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1. INTRODUCTION

It is well known that the Cooper pairing of fermions caused by the kinematic [1], exchange [2, 3], and spin-fluctuation mechanisms considered in the Hubbard [4–6], t–J [2, 3, 7], or t–J* [8–10] models is suppressed by the intersite Coulomb repulsion V of charge carriers located at the neighboring sites. This effect is most pronounced in the d channel [11] and the Cooper instability disappears completely at $V \sim 1$–2 eV [12]. In the case of the s-wave superconducting phase, the Cooper pairing still exists at such values of V [4–6] owing to the stronger kinematic mechanism [1]. There appears a discrepancy between the theory and experiment: in theory, the Coulomb repulsion suppresses the d-wave superconducting phase actually observed in experiment but preserves the s-wave phase, which is not observed in experiments. This limits the potentialities of the mentioned theories in the description of high-T_c superconductors.

In this work, we demonstrate for the first time that taking into account the actual structure of the CuO_2 plane described by the Emery model [13, 14] eliminates the aforementioned discrepancy. In our theory, the Fourier transform of the Coulomb repulsion potential for oxygen holes located at the neighboring sites falls out from the integral equation for the superconducting d-wave phase owing to the symmetry characteristic of this potential. As a result, the $d_{x^2-y^2}$ phase remains stable in the presence of the strong repulsion of neighboring oxygen holes. At the same time, the self-consistency equation for the superconducting s-wave phase includes the Coulomb contribution and the s-wave phase turns out to be suppressed. Therefore, we not only explain the "survivability" of the d-wave phase but also clarify why the $d_{x^2-y^2}$ pairing rather than s-wave pairing appears in cuprate high-T_c superconductors in spite of the strong coupling limit corresponding to the kinematic pairing mechanism.

2. SPIN-FERMION MODEL

For strong electron correlations, when the on-site Coulomb repulsion energy U_d for holes at one copper ion is large $U_d > \Delta_{pd} \gg t_{pd}$, the Emery model is reduced to the spin-fermion model [15, 16]

$$\hat{H} = \hat{H}_0 + \hat{J} + \hat{V} + \hat{I}, \tag{1}$$

where

$$\hat{H}_0 = \sum_{k\alpha}\Big[\xi_0(k_x)a^\dagger_{k\alpha}a_{k\alpha} + \xi_0(k_y)b^\dagger_{k\alpha}b_{k\alpha} + t_k(a^\dagger_{k\alpha}b_{k\alpha} + b^\dagger_{k\alpha}a_{k\alpha})\Big],$$

$$\hat{J} = \frac{J}{N}\sum_{\substack{f k\alpha\\ \alpha\beta}} e^{if(q-k)} u^{\dagger}_{k\alpha}(\mathbf{S}_f \boldsymbol{\sigma}_{\alpha\beta}) u_{q\beta},$$

$$\hat{V} = V\sum_{f\Delta} \hat{n}_{f+\frac{x}{2}} \hat{n}_{f+\frac{x}{2}+\Delta}, \quad \hat{I} = \frac{I}{2}\sum_{\langle fm\rangle} \mathbf{S}_f \mathbf{S}_m,$$

describing the subsystem of oxygen holes interacting with the spins located at copper ions. Here,

$$\begin{aligned}
\xi_0(k_{x(y)}) &= \varepsilon_p - \mu + \tau(1+\cos k_{x(y)}),\\
t_k &= (2\tau - 4t)\cos\frac{k_x}{2}\cos\frac{k_y}{2},\\
u_{k\beta} &= \cos\frac{k_x}{2} a_{k\beta} + \cos\frac{k_y}{2} b_{k\beta},\\
\tau &= \frac{t_{pd}^2}{\Delta_{pd}}\left(1 - \frac{\Delta_{pd}}{U_d - \Delta_{pd} - 2V_{pd}}\right),\\
J &= \frac{4t_{pd}^2}{(\Delta_{pd}+U_p)}\left(1 + \frac{\Delta_{pd}+U_p}{U_d - \Delta_{pd} - 2V_{pd}}\right),\\
I &= \frac{4t_{pd}^4}{(\Delta_{pd}+V_{pd})^2}\left(\frac{1}{U_d} + \frac{2}{2\Delta_{pd}+U_p}\right).
\end{aligned} \tag{2}$$

The Hamiltonian $\hat{H}_0$ describes the subsystem of oxygen holes in the momentum representation. Operators $a^{\dagger}_{k\alpha}$ ($a_{k\alpha}$) create (annihilate) holes with spin $\alpha = \pm 1/2$ in the oxygen subsystem with p_x orbitals and $b^{\dagger}_{k\alpha}$ ($b_{k\alpha}$) are similar operators in the oxygen subsystem with p_y orbitals. Here, ε_p denotes the unrenormalized on-site energy of holes, μ is the chemical potential of the system, and the holes hopping between oxygens with the rate determined by the parameter t. The exchange coupling between the oxygen subsystem and the subsystem of localized spins is described by the operator $\hat{J}$, $\mathbf{S}_f$ is the vector operator of a spin localized at site f, and $\boldsymbol{\sigma} = (\sigma^x, \sigma^y, \sigma^z)$ is the vector of the Pauli matrices. The Coulomb repulsion of holes located at the nearest-neighbor oxygen sites is described by the operator $\hat{V}$, $\hat{n}_{f+x(y)/2} = \sum_{\sigma} \hat{n}_{f+x(y)/2,\sigma}$ is the operator describing the number of holes at the oxygen ion located at site $f + x(y)/2$, $\mathbf{x} = (1, 0)$ and $\mathbf{y} = (0, 1)$ are the basis vectors expressed in units of the lattice constant, and the vector Δ connects the nearest oxygen ions. The last term in the Hamiltonian describes the superexchange interaction between the nearest localized spins, the magnitude of which is determined by the matrix element I.

Further on, we use the well-established values of the involved parameters [12, 17, 18]: $t_{pd} = 1.3$ eV, $\Delta_{pd} = 3.6$ eV, $U_d = 10.5$ eV, $U_p = 4$ eV, $V_{pd} = 1.2$ eV, and $V = 1$–2 eV. For these parameters, the exchange interaction is $I = 0.136$ eV (1570 K), which is in good agreement with the data on high-T_c cuprate superconductors [18]. For the hopping integral, we use the value $t = 0.1$ eV.

It is important that the exchange coupling constant between the localized and itinerant spins calculated according to Eq. (2) is large, namely, $J = 2.4$ eV $\gg \tau \approx 0.1$ eV. Therefore, in the description of the holes at oxygen sites, it is necessary to take into account their strong coupling with the subsystem of spins located at copper ions. This problem is solved using the following basis set of operators [19, 20]:

$$a_{k\alpha}, \quad b_{k\alpha}, \quad L_{k\alpha} = \frac{1}{N}\sum_{fq\beta} e^{if(q-k)}(\mathbf{S}_f \boldsymbol{\sigma}_{\alpha\beta}) u_{q\beta}, \tag{3}$$

where the third operator "entangles" the spin and fermion subsystems.

3. EQUATIONS FOR THE NORMAL AND ANOMALOUS GREEN'S FUNCTIONS

For consideration of the conditions for the Cooper instability, basis (3) is supplemented by the operators ($\bar{\alpha} = -\alpha$)

$$a^{\dagger}_{-k\bar{\alpha}}, \quad b^{\dagger}_{-k\bar{\alpha}}, \quad L^{\dagger}_{-k\bar{\alpha}}. \tag{4}$$

The set of equations for the normal (G_{ij}) and anomalous (F_{ij}) Green's functions obtained by the method described in [21, 22] can be represented in the form ($j = 1, 2, 3$)

$$\begin{aligned}
(\omega - \xi_x)G_{1j} &= \delta_{1j} + t_k G_{2j} + J_x G_{3j} + \Delta_{1k} F_{2j},\\
(\omega - \xi_y)G_{2j} &= \delta_{2j} + t_k G_{1j} + J_y G_{3j} + \Delta_{2k} F_{1j},\\
(\omega - \xi_3)G_{3j} &= \delta_{3j} K_k\\
&\quad + (J_x G_{1j} + J_y G_{2j})K_k + \Delta_{3k} F_{3j},\\
(\omega + \xi_x)F_{1j} &= \Delta^{*}_{2k} G_{2j} - t_k F_{2j} - J_x F_{3j},\\
(\omega + \xi_y)F_{2j} &= \Delta^{*}_{1k} G_{1j} - t_k F_{1j} - J_y F_{3j},\\
(\omega + \xi_3)F_{3j} &= \Delta^{*}_{3k} G_{3j} - (J_x F_{1j} + J_y F_{2j})K_k.
\end{aligned} \tag{5}$$

Here,

$$G_{11} = \left\langle\!\left\langle a_{k\uparrow} | a^{\dagger}_{k\uparrow}\right\rangle\!\right\rangle, \quad G_{21} = \left\langle\!\left\langle b_{k\uparrow} | a^{\dagger}_{k\uparrow}\right\rangle\!\right\rangle,$$
$$G_{31} = \left\langle\!\left\langle L_{k\uparrow} | a^{\dagger}_{k\uparrow}\right\rangle\!\right\rangle.$$

Functions G_{i2} and G_{i3} are determined in a similar way with the only difference that $a^{\dagger}_{k\uparrow}$ is replaced by $b^{\dagger}_{k\uparrow}$ and $L^{\dagger}_{k\uparrow}$, respectively. The anomalous Green's functions can be defined as

$$F_{11} = \left\langle\!\left\langle a^{\dagger}_{-k\downarrow} | a^{\dagger}_{k\uparrow}\right\rangle\!\right\rangle, \quad F_{21} = \left\langle\!\left\langle b^{\dagger}_{-k\downarrow} | a^{\dagger}_{k\uparrow}\right\rangle\!\right\rangle,$$
$$F_{31} = \left\langle\!\left\langle L^{\dagger}_{-k\downarrow} | a^{\dagger}_{k\uparrow}\right\rangle\!\right\rangle.$$

For F_{i2} and F_{i3}, we use the same type of notation according to the second subscript. The functions involved in (5) are determined by the expressions

$$\begin{aligned}\xi_{x(y)} &= \xi_0(k_x(y)) + 4n_pV,\\ J_{x(y)} &= J\cos\frac{k_x(y)}{2}, \quad K_k = 3/4 + C_1\gamma_{1k},\\ \xi_3 &= \varepsilon_p - \mu - 2t + 5\tau/2 - J + n_pV\\ &+ [(\tau - 2t)(C_1\gamma_{1k} + C_2\gamma_{2k}) + \tau(C_1\gamma_{1k} + C_3\gamma_{3k})/2\\ &+ JC_1(1 - 4\gamma_{1k})/4 + IC_1(\gamma_{1k} - 4)]K_k^{-1}.\end{aligned} \tag{6}$$

Here, n_p denotes the hole density per oxygen ion and $\gamma_{1k} = (\cos k_x + \cos k_y)/2$, $\gamma_{2k} = \cos k_x \cos k_y$, and $\gamma_{3k} = (\cos 2k_x + \cos 2k_y)/2$ are invariants for the square lattice. In the course of deriving (5), we assume that the state of localized moments corresponds to the quantum spin liquid. In this case, $\langle S_f^x\rangle = \langle S_f^y\rangle = \langle S_f^z\rangle = 0$ and the correlation functions $C_j = \langle \mathbf{S}_0\mathbf{S}_{r_j}\rangle$ satisfy the relations

$$C_j = 3\langle S_0^x S_{r_j}^x\rangle = 3\langle S_0^y S_{r_j}^y\rangle = 3\langle S_0^z S_{r_j}^z\rangle, \tag{7}$$

where r_j is the position of a copper ion within the jth coordination sphere.

From (5), it follows that the spectrum of the Fermi excitations in the normal phase is determined by the solution of the dispersion equation

$$\begin{aligned}\det_k(\omega) &= (\omega - \xi_x)(\omega - \xi_y)(\omega - \xi_3)\\ &- 2J_xJ_yt_kK_k - (\omega - \xi_y)J_x^2K_k - (\omega - \xi_x)J_y^2K_k\\ &- (\omega - \xi_3)t_k^2 = 0\end{aligned} \tag{8}$$

and is characterized by three branches, ϵ_{1k}, ϵ_{2k}, and ϵ_{3k} [23]. The ϵ_{1k} branch with the minimum at a point close to $(\pi/2, \pi/2)$ arises owing to the strong spin-fermion coupling, which initiates both the exchange interaction between a hole and the nearest-neighbor copper ions and the spin-correlated hoppings. Both upper bands, ϵ_{2k} and ϵ_{3k}, are separated by an appreciable gap from the lower band ϵ_{1k}; therefore, the dynamics of holes when their density n_p is low is determined by the characteristics of the lower band ϵ_{1k}.

The introduced order parameters $\Delta_{j,k}$ are related to the anomalous averages by the following expressions ($C_{1x(1y)} = C_1\cos^2(q_{x(y)}/2)$):

$$\begin{aligned}\Delta_{1k} &= -\frac{4V}{N}\sum_q \phi_{k-q}\langle a_{q\uparrow}b_{-q\downarrow}\rangle,\\ \phi_k &= \cos\frac{k_x}{2}\cos\frac{k_y}{2},\\ \Delta_{2k} &= -\frac{4V}{N}\sum_q \phi_{k-q}\langle b_{q\uparrow}a_{-q\downarrow}\rangle,\\ \Delta_{3k} &= \frac{1}{N}\sum_q\Big\{I_{k-q}\big[\langle L_{q\uparrow}L_{-q\downarrow}\rangle\\ &- C_{1x}\langle a_{q\uparrow}a_{-q\downarrow}\rangle - C_{1y}\langle b_{q\uparrow}b_{-q\downarrow}\rangle\big]K_k^{-1}\\ &+ (\tilde{V}_k - C_1I_{k-q}K_k^{-1})\phi_q\big(\langle a_{q\uparrow}b_{-q\downarrow}\rangle + \langle b_{q\uparrow}a_{-q\downarrow}\rangle\big)\Big\}.\end{aligned} \tag{9}$$

Here, $\tilde{V}_k = V[1 + (C_1\gamma_{1k} + C_2\gamma_{2k})K_k^{-1}]$ and $I_k = 4I\gamma_{1k}$.

4. SET OF EQUATIONS FOR THE SUPERCONDUCTING ORDER PARAMETERS

To find the conditions for appearance of the Cooper instability, we write the anomalous Green's functions in terms of the Δ_{jk}^* parameters in the linear approximation

$$F_{nm}(k,\omega) = \sum_{j=1}^{3} S_{nm}^{(j)}(k,\omega)\Delta_{jk}^*/\mathrm{Det}_k(\omega), \tag{10}$$

where

$$\begin{aligned}&\mathrm{Det}_k(\omega) = -\det_k(\omega)\det_k(-\omega),\\ &S_{12}^{(1)}(k,\omega) = S_{21}^{(2)}(k,\omega) = Q_3(k,-\omega)Q_3(k,\omega),\\ &S_{12}^{(2)}(k,\omega) = Q_{3y}(k,-\omega)Q_{3x}(k,\omega),\\ &S_{12}^{(3)}(k,\omega) = K_kQ_y(k,-\omega)Q_x(k,\omega),\\ &S_{21}^{(1)}(k,\omega) = S_{12}^{(2)}(k,-\omega), \quad S_{21}^{(3)}(k,\omega) = S_{12}^{(3)}(k,-\omega),\\ &S_{11}^{(1)}(k,\omega) = Q_3(k,-\omega)Q_{3y}(k,\omega),\\ &S_{33}^{(2)}(k,\omega) = K_kS_{12}^{(3)}(k,\omega),\\ &S_{11}^{(2)}(k,\omega) = S_{11}^{(1)}(k,-\omega),\\ &S_{11}^{(3)}(k,\omega) = K_kQ_y(k,-\omega)Q_y(k,\omega),\\ &S_{22}^{(1)}(k,\omega) = Q_{3x}(k,-\omega)Q_3(k,\omega),\\ &S_{33}^{(1)}(k,\omega) = K_kS_{21}^{(3)}(k,\omega),\\ &S_{22}^{(2)}(k,\omega) = S_{22}^{(1)}(k,-\omega),\\ &S_{22}^{(3)}(k,\omega) = K_kQ_x(k,-\omega)Q_x(k,\omega),\\ &S_{33}^{(3)}(k,\omega) = K_kQ_{xy}(k,-\omega)Q_{xy}(k,\omega).\end{aligned}$$

The functions used here are defined as follows:

$$\begin{aligned}Q_{x(y)}(k,\omega) &= (\omega - \xi_{x(y)})J_{y(x)} + t_kJ_{x(y)},\\ Q_3(k,\omega) &= (\omega - \xi_3)t_k + J_xJ_yK_k,\\ Q_{3x(3y)}(k,\omega) &= (\omega - \xi_3)(\omega - \xi_{x(y)}) - J_{x(y)}^2K_k,\\ Q_{xy}(k,\omega) &= (\omega - \xi_x)(\omega - \xi_y) - t_k^2.\end{aligned} \tag{11}$$

Then, we use the spectral theorem [24], find the expressions for the anomalous averages, and eventually arrive at the closed set of uniform integral equations for the superconducting order parameters

$$\Delta_{1k}^* = \frac{4V}{N}\sum_{jq}\phi_{k-q}M_{21}^{(j)}(q)\Delta_{jq}^*,$$
$$\Delta_{2k}^* = \frac{4V}{N}\sum_{jq}\phi_{k-q}M_{12}^{(j)}(q)\Delta_{jq}^*,$$
$$\Delta_{3k}^* = \frac{1}{N} \times \sum_{jq}\left\{\frac{I_{k-q}}{K_k}\left[C_{1x}M_{11}^{(j)}(q) + C_{1y}M_{22}^{(j)}(q) - M_{33}^{(j)}(q)\right]\right.$$
$$\left. + (I_{k-q}C_1K_k^{-1} - \tilde{V}_k)\phi_q\left[M_{12}^{(j)}(q) + M_{21}^{(j)}(q)\right]\right\}\Delta_{jq}^*, \quad (12)$$

where

$$M_{nm}^{(j)}(q) = \frac{S_{nm}^{(j)}(q, E_{1q}) + S_{nm}^{(j)}(q, -E_{1q})}{4E_{1q}(E_{1q}^2 - E_{2q}^2)(E_{1q}^2 - E_{3q}^2)}\tanh\left(\frac{E_{1q}}{2T}\right).$$

Set of equations (12) is used to determine the critical temperature of the transition to the superconducting phase having a given type of symmetry.

5. TEMPERATURE OF THE TRANSITION TO THE SUPERCONDUCTING $d_{x^2-y^2}$ PHASE

For the superconducting phase with the $d_{x^2-y^2}$ symmetry of the order parameter, where

$$\Delta_{3k} = \Delta_0 \cdot (\cos k_x - \cos k_y), \quad (13)$$

it follows from Eqs. (12) that $\Delta_{1k} = 0$ and $\Delta_{2k} = 0$ because the kernels of the integral equations for Δ_{1k}^* and Δ_{2k}^* include the function ϕ_{k-q} and the corresponding integrals with respect to q vanish.

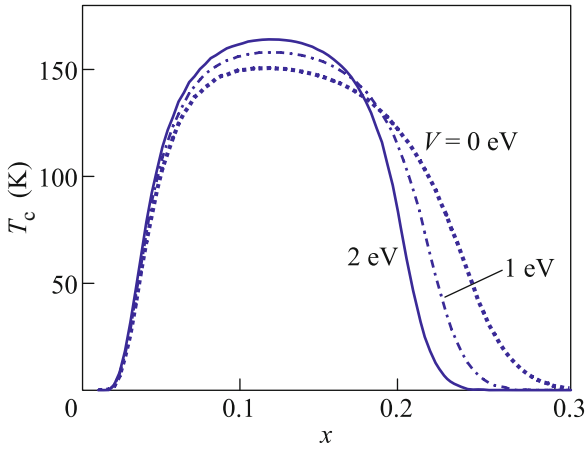
The form of the integral equation for the superconducting order parameter Δ_{3k}^* suggests that the contribution of the intersite Coulomb potential to the kernel of the integral equation vanishes in the case of the d-wave phase. This is due to the symmetry properties of the integrands and manifests itself after the summation over the internal variable. As a result, we arrive at the conclusion that the Coulomb repulsion of holes located at the neighboring oxygen sites does not suppress the superconducting phase with the $d_{x^2-y^2}$ type of the symmetry of the order parameter.

Taking into account the aforementioned facts, we find that the equation determining the dependence of the critical temperature on the charge carrier density has the form

$$1 = \frac{I}{N}\sum_q \frac{(\cos q_x - \cos q_y)^2}{2E_{1q}}\Psi_q \tanh\left(\frac{E_{1q}}{2T_c}\right), \quad (14)$$

where

$$\Psi_q = \left\{S_{33}^{(3)}(q, E_{1q}) - C_{1x}S_{11}^{(3)}(q, E_{1q}) - C_{1y}S_{22}^{(3)}(q, E_{1q}) - C_1\phi_q \times \left[S_{12}^{(3)}(q, E_{1q}) + S_{21}^{(3)}(q, E_{1q})\right]\right\} \times \left[K_q(E_{1q}^2 - E_{2q}^2)(E_{1q}^2 - E_{3q}^2)\right]^{-1}. \quad (15)$$



(Color online) Critical temperature for the transition to the superconducting $d_{x^2-y^2}$ phase versus the hole density at three values of the Coulomb repulsion parameter V.

In the figure, we illustrate the results obtained by solving Eq. (14). The comparison of the presented plots demonstrates that taking into account the intersite Coulomb interaction leads only to slight and nonuniform, with respect to the charge carrier density, modification of the $T_c(x)$ dependence. These insignificant changes are related to the renormalization of the on-site energy of holes due to the Coulomb repulsion at the oxygen ions rather than to the renormalization of the coupling constant.

6. CONCLUSIONS

The main result of our work concerns the answer to the question formulated immediately after theoretical works based on the exchange, kinematic, or spin-fluctuation mechanisms of the Cooper pairing in high-T_c superconductors. The point is that the calculations performed in the framework of the effective model with the simple unit cell (the Hubbard, t–J, and t–J^* models) indicated a strong suppression of superconductivity with the $d_{x^2-y^2}$ symmetry of the order parameter if the Coulomb repulsion between fermions at

neighboring sites was taken into account. In contrast, the superconducting *s*-wave phase initiated by the kinetic mechanism suggested by Zaitsev remains stable but with a significantly reduced critical temperature. There appears a discrepancy between theory and experiment: the experiment exhibited the $d_{x^2-y^2}$-wave superconductivity, whereas it was suppressed in the theory by the Coulomb repulsion of neighboring fermions. At the same time, the theory predicted a relative stability of the superconducting *s*-wave phase with respect to the intersite repulsion of fermions, but this phase has not been observed in experiments.

For the first time, we have shown that the key issue leading to the elimination of such discrepancy between the theory and experiment is the inclusion of the actual structure existing in the CuO_2 plane. It turns out that the Fourier transform of the Coulomb potential vanishes in the set of integral equations self-consistently determining the order parameter of the superconducting phase with the $d_{x^2-y^2}$ symmetry. Therefore, the Coulomb repulsion of holes located at the neighboring oxygen sites does not suppress the Cooper pairing in the *d* channel. On the contrary, the Coulomb potential is involved in the equation for the *s*-wave superconducting phase leading to its suppression. Note that the difference in the contributions of the Coulomb interaction to the conditions of the formation of superconducting phases with different types of symmetry of the order parameters also manifests itself in the Kohn–Luttinger theory of superconductivity [25].

In our case, the spatial separation plays the key role; i.e., two types of oxygen orbitals spatially separated from the spins at copper ions are taken into account. Hence, it becomes evident that the theories of high-T_c superconductivity based on the models using the lattices with a simple unit cell instead of the actual crystal structure are not applicable for the adequate theoretical treatment of the specific features characteristic of cuprate superconductors.

In conclusion, let us focus on the "symmetry cause" responsible for the absence of such contribution to the *d* channel of the Cooper pairing, which is related to the Coulomb repulsion of holes located at the nearest-neighbor oxygen ions. In conventional superconductors, the contribution of the Coulomb potential is renormalized owing to the electron–phonon interaction, whereas in high-T_c superconductors, the elimination of the Coulomb repulsion for the $d_{x^2-y^2}$ phase occurs as a result of the complexity of the unit cell and the specific features of the Fourier transform of the Coulomb potential. This suggests a concept important for the actual implementation of the task-oriented search for novel materials with the high-T_c superconducting phase. Such materials should have a quite complicated unit cell. Moreover, their crystal lattice should have the structure for which the Fourier transform of the Coulomb repulsion between fermions located at the nearest-neighbor sites has the symmetry leading to the vanishing of the contribution to the integral equation for the superconducting order parameters. Just this situation takes place in cuprate superconductors.

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