ELECTRONIC PROPERTIES
OF SOLID

Isotope Effect in the Model of Strongly Correlated Electrons with the Magnetic and Phonon Superconducting Pairing Mechanisms

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Abstract—Peculiarities of the temperature isotope effect in a BCS-type theory describing the exchange and phonon mechanisms of Cooper pairing in a system of strongly correlated electrons are considered. The electron–phonon interaction constant is determined from the fitting of the calculated value of the isotope-effect index to the observed value with the parameters of $La_{2-x}Sr_xCuO_4$ obtained from ab initio calculations. The value of this constant indicates that the contribution from the traditional pairing mechanism to the superconducting state is of the same order of magnitude as the contribution from the magnetic mechanism.

1. INTRODUCTION

The characteristic features of cuprates, such as high superconducting transition temperatures, d-type symmetry of the order parameter, and strong electron correlations in the system, put these materials in a separate class of superconductors and initiated the search for new Cooper pairing mechanisms. The question whether the electron–phonon interaction could ensure high superconducting transition temperatures was widely discussed, and opposite opinions were expressed on this topic [1−3]. Peculiarities of the crystal structure [2] and experimental studies [4, 5] confirm that the electron–phonon interaction in the superconducting states of cuprates is significant, but they do not indicate that this interaction plays a leading role. It is well known that analysis of the isotopic shift of the superconducting transition temperature has played a key role in interpreting the origin of superconductivity in normal metals. In high-temperature superconductors (HTSCs), for which the index of isotope effect strongly depends on the doping level [6−8], the situation is more complicated. As a matter of fact, isotope-effect index α_o in the optimal doping range is minimal for most cuprates with oxygen substitution; moreover, its value is an order of magnitude smaller than the value predicted in the Bardeen–Cooper–Schrieffer (BCS) theory [9]. At the same time, an increase in the value of α_o is observed in weakly doped and overdoped samples; in some systems, the isotope-effect index in the low-doping region considerably exceeds the BCS theory value (i.e., $\alpha_{BCS} = 0.5$). Such

a behavior of the isotope shift constant complicates unambiguous interpretation of the role of the electron–phonon interaction in the superconducting state of cuprates and necessitates analysis of the isotope effect using various approaches. Here, we analyze the isotope effect using a model combining the magnetic and traditional phonon pairing mechanisms in the system of strongly correlated electrons.

2. THEORY OF THE SUPERCONDUCTING STATE TAKING INTO ACCOUNT STRONG ELECTRON CORRELATIONS

It is well known that in a correct description of HTSC systems, strong electron correlations must be taken into account since these correlations control strong changes in physical properties of cuprates upon doping from antiferromagnetic Mott–Hubbard dielectric to Fermi-liquid metals. Several approaches to this problem have been developed. In analyzing the superconducting state in cuprates, the exclusion of two-particle states should exactly be taken into account at all stages of calculation in the representation of Hubbard X-operators evaluating Green's functions using the diagrammatic technique [10] or uncoupling of the corresponding equations of motion [11]. An analogous prohibition imposed on doubly occupied states also exists in the Monte Carlo variational method for projection wavefunctions [12], in the theories of resonating valence bonds [13] and of

renormalized mean field constructed in the Gutzwiller approximation [14, 15].

Here, we develop a generalized tight-binding method [16], in which a transition from the realistic multiband $p-d$ model of cuprates to the effective Hubbard model is carried out. Hubbard operators are constructed on the eigenstates of the Hamiltonian of the CuO_6 cluster, which are obtained as a result of its exact diagonalization: $X_f^{pq} = |p\rangle\langle q|$. The upper and lower bands of Hubbard fermions describe excitations of one-particle and singlet two-particle states, respectively. The elimination of interband transitions in perturbation theory [17, 18] leads to the effective low-energy $t-J^*$ model. The Hamiltonian of the hole-doped $La_{2-x}Sr_xCuO_4$ system has the form

$$H_{tJ^*} = H_{tJ} + H_{(3)},$$

$$H_{tJ} = \sum_{f\sigma}(\varepsilon - \mu)X_f^{\sigma\sigma}$$

$$+ \sum_f(2\varepsilon - 2\mu + U_{\text{eff}})X_f^{22} + \sum_{fg\sigma}t_{fg}X_f^{2\bar{\sigma}}X_g^{\bar{\sigma}2}$$

$$+ \frac{1}{2}\sum_{fg\sigma}J_{fg}(X_f^{\sigma\bar{\sigma}}X_g^{\bar{\sigma}\sigma} - X_f^{\sigma\sigma}X_g^{\bar{\sigma}\bar{\sigma}}),$$

$$H_{(3)} = \sum_{fmg\sigma}\frac{\tilde{t}_{fm}\tilde{t}_{mg}}{U_{\text{eff}}}(X_f^{2\bar{\sigma}}X_m^{\sigma\sigma}X_g^{\bar{\sigma}2} - X_f^{2\sigma}X_m^{\sigma\sigma}X_g^{\bar{\sigma}2}).$$

Here, μ is the chemical potential of the system, t_{fg} and $\tilde{t}_{fg}$ are the amplitudes of intraband and interband jumps, U_{eff} is the dielectric gap with the charge transfer energy, $J_{fg} = t_{fg}^2/U_{\text{eff}}$ is the exchange interaction parameter, and $\bar{\sigma} \equiv -\sigma$. Three-center correlated jumps $H_{(3)}$ have the same order of magnitude as the exchange integral and, according to [19], must be taken into account in the theory of superconductivity.

In analyzing the superconducting d-wave state, passage from the Hubbard model to the low-energy $t-J^*$ Hamiltonian indicates disregard for the retarded spin-fluctuation pairing mechanism [11, 20]. Thus, we confine our analysis to instantaneous exchange interaction in the $t-J^*$ model. It was shown earlier [11] in the self-consistent Born approximation for the mass operator that the influence of retardation effects on d-type superconducting pairing in the Hubbard model is insignificant, which makes it possible to approximate it by instantaneous interaction. At the same time, the relation between retarded and nonretarded contributions to the pairing d-type interaction in real compounds requires further investigation [21, 22].

The complete Hamiltonian of the system also includes the electron–phonon interaction:

$$H_{\text{tot}} = H_{tJ^*} + H_{\text{epi}}. \tag{1}$$

In the case of strong electron correlations, the Fourier transform of the effective interaction of electrons via the exchange of virtual phonons can be written in the form [23]

$$H_{\text{epi}} = \sum_{\mathbf{kk'q}}\sum_{\sigma\sigma'}V_{\mathbf{kk'q}}X_{\mathbf{k+q}}^{2,\bar{\sigma}}X_{\mathbf{k'-q}}^{2,\bar{\sigma}'}X_{\mathbf{k'}}^{\bar{\sigma}',2}X_{\mathbf{k}}^{\bar{\sigma},2}.$$

Disregarding the retardation effects, we can write the effective electron–phonon interaction parameter in a simpler form:

$$V_{\mathbf{kk'q}} = -\sum_\nu g_\nu(\mathbf{k},\mathbf{q})g_\nu(\mathbf{k'},-\mathbf{q})/\omega_{\mathbf{q},\nu}.$$

Here, $g_\nu(\mathbf{k},\mathbf{q})$ is the matrix element of an electron with initial momentum $\mathbf{k}$ and a phonon of the ν mode with momentum $\mathbf{q}$ and oscillation frequency $\omega_{\mathbf{q},\nu}$.

In a BCS-type theory with Hamiltonian (1), the superconducting d-wave order parameter obtained in the generalized Hartree–Fock approximation using the irreducible linear operator method [24] has the form [25]

$$\Delta_{\mathbf{k}} = \frac{2\varphi_{\mathbf{k}}}{N}\sum_{\mathbf{q}}\lambda_{\text{tot}}\frac{2\Delta_{\mathbf{q}}\varphi_{\mathbf{q}}}{\xi_{\mathbf{q}} - \mu}\tanh\left(\frac{\xi_{\mathbf{q}} - \mu}{2k_BT}\right). \tag{2}$$

Here, k_B is the Boltzmann constant, T is the temperature, $\xi'_{\mathbf{q}}$ is the superconducting phase dispersion, μ is the chemical potential of the system, $\varphi_{\mathbf{k}} = (\cos k_x a - \cos k_y a)/2$ is the angular part of order parameter $\Delta_{\mathbf{k}} = \Delta_0\varphi_{\mathbf{k}}$, and a is the lattice constant. The total coupling constant describes the sum of the exchange and phonon pairing mechanisms:

$$\lambda_{\text{tot}} = \frac{1-x}{2}J + \lambda_{\text{ph}}\theta(|\xi_{\mathbf{q}} - \mu| - \omega_D),$$

where x is the concentration of doped holes and the θ-function confines as usual the phonon contribution to a narrow layer on the order of Debye frequency ω_D near the Fermi surface. An equation with an analogous structure was obtained earlier for the magnetic pairing mechanism in the theory of resonating valence bonds [13] as well as by the methods of decoupling of two-time Green's functions [26] and diagrammatic technique [10].

Let us consider in greater detail the electron–phonon coupling constant $\lambda_{\text{ph}} = f(x)G$. Dimensionless parameter $f(x) = (1 + x)(3 + x)/8 - 3C_{01}/4$ describes its concentration dependence originating, as in the case of the exchange mechanism, from the peculiar statistics of Hubbard fermions. In contrast to bands of free electrons, Hubbard subbands contain an odd number of states per atom, which depends on doped carrier concentration x in terms of their total number $n_h = 1 + x$. The emergence of spin correlation functions $C_{01} = 2\langle S_0^z S_1^z\rangle$ indicates interference of the magnetic and phonon pairing mechanisms. We believe that the magnetic system is in the state of an isotropic spin

liquid for which the mean value of any spin component is zero: $\langle S^\alpha \rangle = 0$, $\alpha = x, y, z$. Interatomic static correlations functions differ from zero ($\langle S_0^\alpha S_n^\alpha \rangle \neq 0$) and are identical for all spin components so that $2\langle S_0^z S_n^z \rangle = \langle S_0^+ S_n^- \rangle$. Here, n is the number of the coordination sphere; i.e., C_{01} is the correlation function for the nearest neighbors. In view of the antiferromagnetic nature of correlations, $C_{01} < 0$, so that the interference term with C_{01} increases the total phonon coupling constant.

In deriving the equation for the order parameter, we considered only three vibrational modes for oxygen (buckling, plane breathing, and apical breathing modes). The ab initio calculations of the electronic structure and the electron–phonon interaction in $La_{2-x}Sr_xCuO_4$ demonstrated that the main contribution to the self-energy of electrons is due to the interaction of carriers with just these vibrations [27]. It turns out that the momentum dependence of the matrix elements of the electron–phonon interaction is significant for the d-wave pairing [25]. For an isotropic pairing of the s-type, the contributions from all phonon modes are summed, while the coupling constant in the $d_{x^2-y^2}$-channel can be suppressed by certain vibrations. This is due to the fact that the interaction between an electron and a phonon with large transferred momenta $\mathbf{q}$ transfers the electron to a region of the Fermi surface with another phase of the order parameter, thus breaking the superconducting pair. Conversely, oscillations for which the matrix element has the maximal value for small transferred momenta increase the coupling constant. This fact reflects the competition of the buckling and breathing modes in the expression for parameter G proportional to the intensities of the matrix elements of the electron–phonon interaction:

$$G = \frac{V_{\text{buckl}}^2}{\omega_{\text{buckl}}} - \frac{V_{\text{breath}}^2}{\omega_{\text{breath}}}. \tag{3}$$

An analogous conclusion was drawn earlier in the random phase approximation in the 2D Hubbard model [28], fluctuating exchange (FLEX) for the Eliashberg equation [22], by the method of cluster perturbation theory [30] and functional renorm-group method [31].

It should also be noted that zero contribution from the apical breathing mode to the coupling constant, which was also noted in [28], follows from the orthogonality of the electron momentum lying in the CuO plane and the phonon wavevector. The strong electron–phonon interaction for this mode associated with the Coulomb interaction weakly screened along the c axis is confirmed in experiments, in which colossal expansion of lanthanum cuprate films induced by photodoping was observed [5]. At the same time, analysis of the isotope effect in experiments [32, 33] with position-sensitive substitution $O^{16} \longrightarrow O^{18}$ indicates

that apical oxygen vibrations make zero contribution to the isotope effect and, hence, to the emergence of superconductivity.

The expression for normal phase dispersion $\xi_\mathbf{k}$ appearing in Eq. (2) takes into account the short-range magnetic order in the system:

$$\xi_\mathbf{k} = \varepsilon_0 + U_{\text{eff}} - \mu + \frac{1+x}{2}t_\mathbf{k} + \frac{1-x^2}{4}\frac{\tilde{t}_\mathbf{k}^2}{U_{\text{eff}}}$$

$$+ \frac{1}{N}\sum_\mathbf{q}\left(t_{\mathbf{k}-\mathbf{q}} - \frac{1-x}{2}J_\mathbf{q} + \frac{1-x}{2}\frac{\tilde{t}_{\mathbf{k}-\mathbf{q}}^2}{U_{\text{eff}}}\right.$$

$$\left. - (1+x)\frac{\tilde{t}_\mathbf{k}\tilde{t}_{\mathbf{k}-\mathbf{q}}}{U_{\text{eff}}}\right)\frac{2}{1+x}\frac{3}{2}C_\mathbf{q} + \frac{1}{N}$$

$$\times \sum_\mathbf{q}\left(t_\mathbf{q} - \frac{1-x}{2}J_{\mathbf{k}-\mathbf{q}} - x\frac{\tilde{t}_\mathbf{q}^2}{U_{\text{eff}}}\right.$$

$$\left. - (1+x)\frac{\tilde{t}_\mathbf{k}\tilde{t}_{\mathbf{k}-\mathbf{q}}}{U_{\text{eff}}}\right)\frac{2}{1+x}K_\mathbf{q}.$$

Here, $C_\mathbf{q}$ and $K_\mathbf{q}$ are the spin and kinematic static correlation functions describing the renormalization of the quasiparticle spectrum beyond the Hubbard-I approximation. A similar equation for the lower Hubbard band was considered in [34], in which a considerable modification of the quasiparticle spectrum in the $t–J^*$ model by magnetic fluctuations was observed. The parameters of the electronic structure of $La_{2-x}Sr_xCuO_4$ were determined in [35] by projecting the band structure determined in the local-density approximation onto the basis of Wannier functions followed by the passage from the $p–d$ to the $t–J^*$ model using the generalized method of strong coupling. Self-consistent calculation of the correlation functions for the given parameters was performed using the approach developed in [34, 36] by the authors of [37], who studied the rearrangement of the Fermi surface with doping in the normal phase of HTSC cuprates.

3. ISOTOPE EFFECT

In analyzing the isotope effect, we pass from expression (2) to the equation for the superconducting transition temperature T_c, and further, in accordance with the definition of $\alpha_o = -d\ln(T_c)/d\ln(M_o)$, we obtain

$$\alpha_o = \frac{\dfrac{\omega_\mathrm{D}}{N}\sum_\mathbf{q}\lambda_{\text{ph}}\delta(|\xi_\mathbf{q} - \mu| - \omega_\mathrm{D})\dfrac{4\varphi_\mathbf{q}^2}{\xi_\mathbf{q}}\tanh\left(\dfrac{\xi_\mathbf{q} - \mu}{2k_\mathrm{B}T_c}\right)}{\dfrac{1}{N}\sum_\mathbf{q}\lambda_{\text{tot}}(\mathbf{q})\dfrac{4\varphi_\mathbf{q}^2}{T_c}\cosh^{-2}\left(\dfrac{\xi_\mathbf{q} - \mu}{2k_\mathrm{B}T_c}\right)}. \tag{4}$$

It follows from this equation that the sign of the isotope-effect index depends on the effective electron–phonon interaction parameter G, which is considered here as a free parameter in contrast to [25]. Due to the
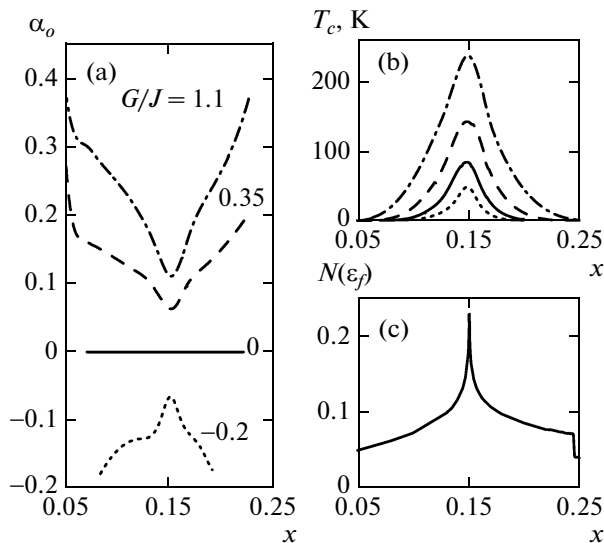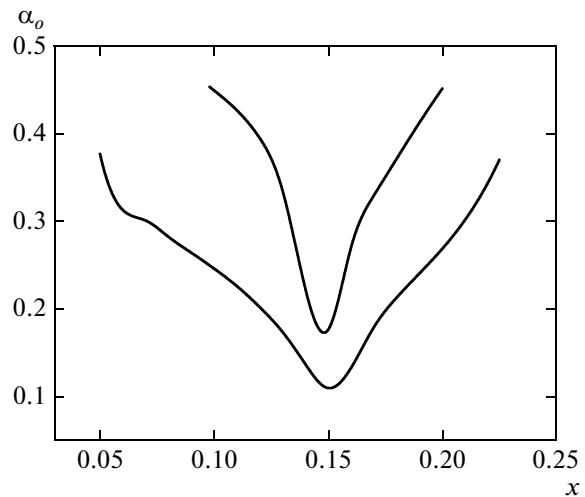
Fig. 1. Concentration dependence of (a) isotope-effect index, (b) superconducting transition temperature, and (c) density of states at the Fermi level. Temperature curves correspond to case (a).

Fig. 2. Isotope-effect index for purely phonon (upper curve) and the sum of the phonon and exchange mechanisms of pairing (lower curve) for $G/J = 1.1$.

competition of various vibrational contributions (3), this parameter can be either positive (which corresponds to the sum of the exchange and phonon pairing mechanisms) or negative (when the superconducting state of the electron–phonon interaction is suppressed). For any $G > 0$, isotope-effect index α_o is also positive; with decreasing phonon contribution to the total coupling constant, the value of α_o decreases and becomes negative for $G < 0$ in the entire range of the solutions (Fig. 1a).

The situation observed in cuprates is described by curves with $G > 0$, for which the concentration dependence $\alpha_o(x)$ is in qualitative agreement with experiment [6−8]. The minimum of the isotope effect corresponds to the maximum of the superconducting transition temperature (Fig. 1b) defined in our approach by the Van Hove singularity in the density of states (Fig. 1c). Note that the value of index $\alpha_o \sim (0.08-0.11)$ characteristic of $La_{2-x}Sr_xCuO_4$ in the optimal doping region is reproduced in calculations with $G/J \sim 1$, which is an indication of the strong electron–phonon coupling. Finally, the stronger increase in the isotope-effect index in the low-doping region for real compounds undergoing a transition from the orthorhombic to the tetragonal phase may be associated with an admixture of an s-symmetry component to the superconducting order parameter of the $d_{x^2-y^2}$-type [38−41]. However, analysis of this problem is beyond the scope of this article, as well as the effects of charge ordering in the $La_{2-x}Sr_xCuO_4$ structure leading to anomalies in properties for $x = 1/8$.

Analysis of Eq. (4) also shows that the addition of any nonphonon mechanism to the traditional mecha-

nism of Cooper pairing leads to a decrease in the isotope-effect index. Figure 2 shows that for a given doping level and $G/J = 1.1$, constant α_o decreases by almost half in the case describing the sum of the phonon and exchange pairing mechanisms as compared to the pure phonon mechanism. In accordance with the above arguments, the contribution of the electron–phonon interaction to superconducting pairing in cuprates in this theory is comparable to the exchange contribution in spite to the weakness of the isotope effect under optimal doping.

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Translated by N. Wadhwa