High Temperature Superconductivity and Charge Segregation in a Model with Strong Long-Range Electron-Phonon and Coulomb Interactions

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An analytical method of studying strong long-range electron-phonon and Coulomb interactions in complex lattices is presented. The method is applied to a perovskite layer with anisotropic coupling of holes to the vibrations of apical atoms. Depending on the relative strength of the polaronic shift, E_p, and the inter-site Coulomb repulsion, V_c, the system is either a polaronic Fermi liquid, $V_c > 1.23\,E_p$, a bipolaronic superconductor, $1.16\,E_p < V_c < 1.23\,E_p$, or a charge segregated insulator, $V_c < 1.16\,E_p$. In the superconducting window, the carriers are mobile bipolarons with a remarkably low effective mass. The model describes the key features of the underdoped superconducting cuprates.

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There is clear experimental [1–5] and theoretical [6–15] evidence for strong electron-phonon (el-ph) interaction in high-T_c cuprates. Electron correlations are also important in shaping the Mott-Hubbard insulating state of parent undoped compounds [16]. The theory of high-T_c cuprates must treat both interactions on equal footing as was suggested some time ago [6]. In recent years many publications addressed the fundamental problem of competing el-ph and Coulomb interactions in the framework of the Holstein-Hubbard model [11–15] where both interactions are short-range (on-site). The mass of bipolaronic carriers in this model is very large and the critical temperature is suppressed down to a kelvin scale. However, in the cuprates the screening is poor so that the el-ph interaction necessarily has to be long range. Motivated by this fact, we have proposed that a long range Fröhlich, rather than short range Holstein, interaction should be the adequate model for the cuprates [17,18]. A small polaron with the Fröhlich interaction was discussed long time ago [19]. Analytical [17] and exact Monte-Carlo [18] studies of the simple chain and plane models with a long-range el-ph coupling revealed a several order lower effective mass of this polaron than that of the small Holstein polaron. Later the polaron and bipolaron cases of the chain model were analyzed in more detail in Refs. [20] and [21] confirming low masses of both types of carriers. Qualitatively, a long-range el-ph interaction results in a lighter mass because the extended lattice deformation changes gradually as the carrier tunnels through the lattice.

In this Letter we study a realistic multi-polaron model of the copper-oxygen perovskite layer, a major structural unit of the HTSC compounds. The model includes the infinite on-site repulsion (Hubbard U term), long-range inter-hole Coulomb repulsion V_c, and long-range Fröhlich interaction between in-plane holes and apical oxygens. We find that within a certain window of V_c the holes form intersite bipolarons with a remarkably low mass. The bipolarons repel and the whole system is a superconductor with a high critical temperature. At large V_c, the system is a polaronic Fermi-liquid, and at small V_c it is a charge segregated insulator.

To deal with the considerable complexity of the model we first describe a theoretical approach that makes the analysis of complex lattices simple in the strong coupling limit. The model Hamiltonian explicitly includes long-range electron-phonon and Coulomb interactions as well as kinetic and deformation energies. An implicitly present infinite Hubbard term prohibits double occupancy and removes the need to distinguish fermionic spins. Introducing spinless fermion, $c_{\mathbf{n}}$, and phonon, $d_{\mathbf{m}\alpha}$, operators the Hamiltonian is written as

$$\begin{aligned} H = & -\sum_{\mathbf{n}\neq\mathbf{n}'}\Big[T(\mathbf{n}-\mathbf{n}')c^\dagger_{\mathbf{n}}c_{\mathbf{n}'} - V_c(\mathbf{n}-\mathbf{n}')c^\dagger_{\mathbf{n}}c_{\mathbf{n}}c^\dagger_{\mathbf{n}'}c_{\mathbf{n}'}\Big] \\ & -\omega\sum_{\mathbf{nm}} g_\alpha(\mathbf{m}-\mathbf{n})(\mathbf{e}_{\mathbf{m}\alpha}\cdot\mathbf{u}_{\mathbf{m}-\mathbf{n}})c^\dagger_{\mathbf{n}}c_{\mathbf{n}}(d^\dagger_{\mathbf{m}\alpha}+d_{\mathbf{m}\alpha}) \\ & +\omega\sum_{\mathbf{m}\alpha}\left(d^\dagger_{\mathbf{m}\alpha}d_{\mathbf{m}\alpha}+\frac{1}{2}\right). \end{aligned} \tag{1}$$

Here $\mathbf{e}_{\mathbf{m}\alpha}$ is the polarization vector of αth vibration coordinate at site $\mathbf{m}$, $\mathbf{u}_{\mathbf{m}-\mathbf{n}} \equiv (\mathbf{m}-\mathbf{n})/|\mathbf{m}-\mathbf{n}|$ is the unit vector in the direction from electron $\mathbf{n}$ to ion $\mathbf{m}$, and $g_\alpha(\mathbf{m}-\mathbf{n})$ is a dimensionless el-ph coupling function. [$g_\alpha(\mathbf{m}-\mathbf{n})$ is proportional to the *force* acting between $\mathbf{m}$ and $\mathbf{n}$.] We assume that all the phonon modes are dispersionless with frequency ω and that the electrons do not interact with displacements of their own atoms, $g_\alpha(0) \equiv 0$. We also use $\hbar = 1$ throughout the paper.

In the limit of strong el-ph interaction it is convenient to perform the Lang-Firsov canonical transformation [22] . Introducing $S = \sum_{\mathbf{mn}\alpha} g_\alpha(\mathbf{m}-\mathbf{n})(\mathbf{e}_{\mathbf{m}\alpha}\cdot$

$\mathbf{u_{m-n}})c^\dagger_{\mathbf{n}}c_{\mathbf{n}}(d^\dagger_{\mathbf{m}\alpha} - d_{\mathbf{m}\alpha})$ one obtains a transformed Hamiltonian without an explicit el-ph term

$$\tilde{H} = e^{-S}He^S = -\sum_{\mathbf{n}\neq\mathbf{n}'}\hat{\sigma}_{\mathbf{nn}'}c^\dagger_{\mathbf{n}}c_{\mathbf{n}'} + \omega\sum_{\mathbf{m}\alpha}\left(d^\dagger_{\mathbf{m}\alpha}d_{\mathbf{m}\alpha} + \frac{1}{2}\right) + \sum_{\mathbf{n}\neq\mathbf{n}'}v(\mathbf{n}-\mathbf{n}')c^\dagger_{\mathbf{n}}c_{\mathbf{n}}c^\dagger_{\mathbf{n}'}c_{\mathbf{n}'} - E_p\sum_{\mathbf{n}}c^\dagger_{\mathbf{n}}c_{\mathbf{n}}. \quad (2)$$

The last term describes the energy which polarons gain due to el-ph interaction. E_p is the familiar polaronic (Franc-Condon) level shift

$$E_p = \omega\sum_{\mathbf{m}\alpha}g^2_\alpha(\mathbf{m}-\mathbf{n})(\mathbf{e_{m}}\alpha\cdot\mathbf{u_{m-n}})^2, \quad (3)$$

which we assume to be independent of $\mathbf{n}$. E_p is a natural measure of the strength of the el-ph interaction. The third term in Eq.(2) is the polaron-polaron interaction:

$$v(\mathbf{n}-\mathbf{n}') = V_c(\mathbf{n}-\mathbf{n}') - V_{\rm pa}(\mathbf{n}-\mathbf{n}'), \quad (4)$$

$$V_{\rm pa}(\mathbf{n}-\mathbf{n}') = 2\omega\sum_{\mathbf{m}\alpha}g_\alpha(\mathbf{m}-\mathbf{n})g_\alpha(\mathbf{m}-\mathbf{n}')\times (\mathbf{e_{m}}\alpha\cdot\mathbf{u_{m-n}})(\mathbf{e_{m}}\alpha\cdot\mathbf{u_{m-n'}}), \quad (5)$$

where $V_{\rm pa}$ is the inter-polaron *attraction* due to joint interaction with the same vibrating atoms. Finally, the first term in Eq.(2) contains a transformed hopping operator $\hat{\sigma}_{\mathbf{nn}'}$:

$$\hat{\sigma}_{\mathbf{nn}'} = T(\mathbf{n}-\mathbf{n}')\exp\left[\sum_{\mathbf{m}\alpha}[g_\alpha(\mathbf{m}-\mathbf{n})(\mathbf{e_{m}}\alpha\cdot\mathbf{u_{m-n}}) - g_\alpha(\mathbf{m}-\mathbf{n}')(\mathbf{e_{m}}\alpha\cdot\mathbf{u_{m-n'}})]\,(d^\dagger_{\mathbf{m}\alpha} - d_{\mathbf{m}\alpha})\right]. \quad (6)$$

At large $E_p/T(\mathbf{n}-\mathbf{n}')$ this term is a perturbation. In the first order of the strong coupling perturbation theory [6], $\hat{\sigma}_{\mathbf{nn}'}$ should be averaged over phonons because there is no coupling between polarons and phonons in the unperturbed Hamiltonian [the last three terms in Eq.(2)]. For temperatures lower than ω, the result is

$$t(\mathbf{n}-\mathbf{n}') \equiv \langle\hat{\sigma}_{\mathbf{nn}'}\rangle_{ph} = T(\mathbf{n}-\mathbf{n}')\exp[-G^2(\mathbf{n}-\mathbf{n}')], \quad (7)$$

$$G^2(\mathbf{n}-\mathbf{n}') = \sum_{\mathbf{m}\alpha}g_\alpha(\mathbf{m}-\mathbf{n})(\mathbf{e_{m}}\alpha\cdot\mathbf{u_{m-n}})\times [g_\alpha(\mathbf{m}-\mathbf{n})(\mathbf{e_{m}}\alpha\cdot\mathbf{u_{m-n}}) - g_\alpha(\mathbf{m}-\mathbf{n}')(\mathbf{e_{m}}\alpha\cdot\mathbf{u_{m-n'}})]. \quad (8)$$

By comparing Eqs.(3), (5), and (8), the mass renormalization exponents can be expressed via E_p and $V_{\rm pa}$ as follows

$$G^2(\mathbf{n}-\mathbf{n}') = \frac{1}{\omega}\left(E_p - \frac{1}{2}V_{\rm pa}(\mathbf{n}-\mathbf{n}')\right). \quad (9)$$

This is the simplest way to calculate G^2 and (bi)polaron masses once the 'static' parameters E_p and $V_{\rm pa}$ are known.

It is easy to see from the above equations that the long-range el-ph interaction increases E_p and $V_{\rm pa}$ but *reduces* G^2 (when measured in natural units of E_p/ω). Thus polarons get tighter and at the same time lighter. Bipolarons form when $V_{\rm pa}$ exceeds V_c and they are relatively light too. We note that the Holstein model is the limiting case with the highest possible $G^2 = E_p/\omega$. In this respect, the Holstein model is *not* a typical el-ph model.

To obtain an analytical description of the multipolaron system we restrict our consideration to the strong coupling case $t \leq |v|$. In this regime the polaron kinetic energy is the smallest energy and thus can be treated as a perturbation. The system is adequately described by a purely polaronic model:

$$H_p = H_0 + H_{\rm pert}, \quad (10)$$

$$H_0 = -E_p\sum_{\mathbf{n}}c^\dagger_{\mathbf{n}}c_{\mathbf{n}} + \sum_{\mathbf{n}\neq\mathbf{n}'}v(\mathbf{n}-\mathbf{n}')c^\dagger_{\mathbf{n}}c_{\mathbf{n}}c^\dagger_{\mathbf{n}'}c_{\mathbf{n}'}, \quad (11)$$

$$H_{\rm pert} = -\sum_{\mathbf{n}\neq\mathbf{n}'}t(\mathbf{n}-\mathbf{n}')c^\dagger_{\mathbf{n}}c_{\mathbf{n}'}. \quad (12)$$

The many-particle ground state of H_0 depends on the sign of the polaron-polaron interaction, the carrier density, and the lattice geometry. Here we consider a two dimensional lattice of ideal octahedra that can be regarded as a simplified model of the copper-oxygen perovskite layer, see Figure 1. The lattice period is $a = 1$ and the distance between the apical sites and the central plane is $h = a/2 = 0.5$. All in-plane atoms, both copper and oxygen, are static but apical oxygens are independent three-dimensional isotropic harmonic oscillators. Because of poor screening the hole-apical interaction is purely Coulombic, $g_\alpha(\mathbf{m}-\mathbf{n}) = \kappa_\alpha/|\mathbf{m}-\mathbf{n}|^2$, $\alpha = x, y, z$. To account for the experimental fact that z-polarized phonons couple to the holes stronger than the others [3] we choose $\kappa_x = \kappa_y = \kappa_z/\sqrt{2}$. The direct hole-hole repulsion is $V_c(\mathbf{n}-\mathbf{n}') = \frac{V_c/\sqrt{2}}{|\mathbf{n}-\mathbf{n}'|}$ so that the repulsion between two holes in the NN configuration is V_c. We also include the bare nearest neighbor (NN) hopping T_{NN}, the next nearest neighbor (NNN) hopping across copper T_{NNN} and the NNN hopping between octahedra T'_{NNN}.

According to Eq.(3), the polaron shift is given by the lattice sum (after summation over polarizations):

$$E_p = 2\kappa_x^2\omega\sum_{\mathbf{m}}\left(\frac{1}{|\mathbf{m}-\mathbf{n}|^4} + \frac{h^2}{|\mathbf{m}-\mathbf{n}|^6}\right) = 31.15\,\kappa_x^2\omega, \quad (13)$$

where the factor 2 accounts for the two layers of apical sites. [For reference, Cartesian coordinates are $\mathbf{n} = (n_x +$

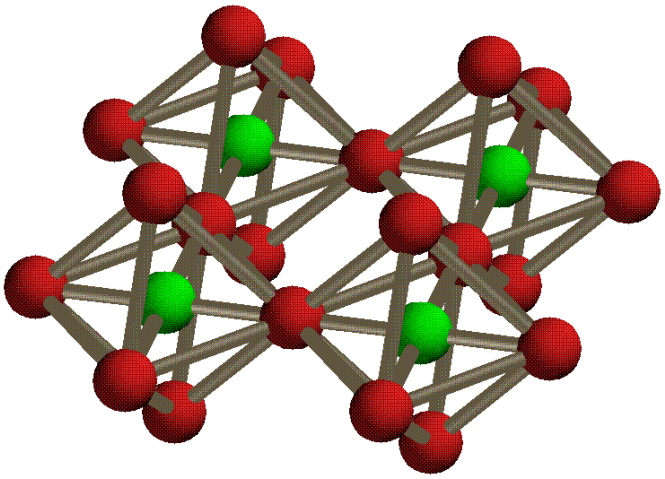

FIG. 1. Four octahedra of the copper-oxygen perovskite layer. Holes reside on the in-plane oxygens but interact with apical oxygens.

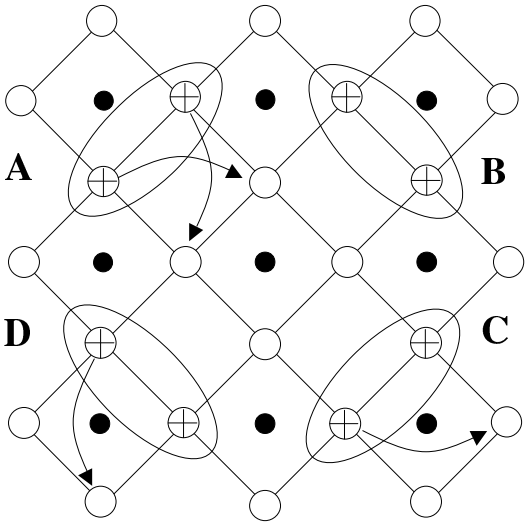

FIG. 2. Top view on the perovskite layer. The apical sites are not shown. The four bipolaron configurations A, B, C, and D all have the same energy. Some possible single-polaron hoppings t' are indicated by arrows. Note that the bipolaron movement is first order in t'.

$1/2, n_y + 1/2, 0)$, $\mathbf{m} = (m_x, m_y, h)$; n_x, n_y, m_x, m_y being integers.] The polaron-polaron attraction is

$$V_{\rm pa}(\mathbf{n}-\mathbf{n}') = 4\omega\kappa_x^2 \sum_{\mathbf{m}} \frac{h^2 + (\mathbf{m}-\mathbf{n}')\cdot(\mathbf{m}-\mathbf{n})}{|\mathbf{m}-\mathbf{n}'|^3|\mathbf{m}-\mathbf{n}|^3}. \quad (14)$$

Performing lattice summations for the NN, NNN, and NNN$'$ configurations one finds $V_{\rm pa} = 1.23\,E_p, 0.80\,E_p$, and $0.82\,E_p$, respectively. Substituting these results in Eqs.(4) and (9) we obtain the full inter-polaron interaction: $v_{NN} = V_c - 1.23\,E_p$, $v_{NNN} = \frac{V_c}{\sqrt{2}} - 0.80\,E_p$, $v'_{NNN} = \frac{V_c}{\sqrt{2}} - 0.82\,E_p$, and the mass renormalization exponents: $G^2_{NN} = 0.38(E_p/\omega)$, $G^2_{NNN} = 0.60(E_p/\omega)$ and $G'^2_{NNN} = 0.59(E_p/\omega)$.

Let us now discuss different regimes of the model. At $V_c > 1.23\,E_p$, no bipolarons are formed and the systems is a polaronic Fermi liquid. The polarons tunnel in the *square* lattice with NN hopping $t = T_{NN}\exp(-0.38E_p/\omega)$ and NNN hopping $t' = T_{NNN}\exp(-0.60E_p/\omega)$. [Since $G^2_{NNN} \approx G'^2_{NNN}$ one can neglect the difference between NNN hoppings within and between the octahedra.] The single polaron spectrum is therefore

$$E_1(\mathbf{k}) = -E_p - 2t'[\cos k_x + \cos k_y] \pm 4t\cos(k_x/2)\cos(k_y/2). \quad (15)$$

The polaron mass is $m^* = 1/(t+2t')$. Since in general $t > t'$, the mass is mostly determined by the NN hopping amplitude t.

If $V_c < 1.23\,E_p$, then intersite NN bipolarons form. The bipolarons tunnel in the plane via four resonating (degenerate) configurations A, B, C, and D, see Figure 2. In the first order in $H_{\rm pert}$ one should retain only these lowest energy configurations and discard all the processes that involve configurations with higher energies. The result of such a projection is the bipolaronic Hamiltonian

$$\begin{aligned} H_b = {} & (V_c - 3.23\,E_p)\sum_{\mathbf{l}}[A^\dagger_{\mathbf{l}}A_{\mathbf{l}} + B^\dagger_{\mathbf{l}}B_{\mathbf{l}} + C^\dagger_{\mathbf{l}}C_{\mathbf{l}} + D^\dagger_{\mathbf{l}}D_{\mathbf{l}}] \\ & - t'\sum_{\mathbf{l}}[A^\dagger_{\mathbf{l}}B_{\mathbf{l}} + B^\dagger_{\mathbf{l}}C_{\mathbf{l}} + C^\dagger_{\mathbf{l}}D_{\mathbf{l}} + D^\dagger_{\mathbf{l}}A_{\mathbf{l}} + \text{h.c.}] \\ & - t'\sum_{\mathbf{n}}[A^\dagger_{\mathbf{l}-\mathbf{x}}B_{\mathbf{l}} + B^\dagger_{\mathbf{l}+\mathbf{y}}C_{\mathbf{l}} \\ & + C^\dagger_{\mathbf{l}+\mathbf{x}}D_{\mathbf{l}} + D^\dagger_{\mathbf{l}-\mathbf{y}}A_{\mathbf{l}} + \text{h.c.}], \end{aligned} \quad (16)$$

where $\mathbf{l}$ numbers octahedra rather than individual sites, $\mathbf{x} = (1,0)$, and $\mathbf{y} = (0,1)$. A Fourier transformation and diagonalization of a 4×4 matrix yields the bipolaron spectrum:

$$E_2(\mathbf{k}) = V_c - 3.23E_p \pm 2t'[\cos(k_x/2) \pm \cos(k_y/2)]. \quad (17)$$

There are four bipolaronic subbands combined in a band of width $8t'$. The effective mass of the lowest band is $m^{**} = 2/t'$. The bipolaron binding energy is $\Delta = 2E_1(0) - E_2(0) = 1.23E_p - V_c - 8t - 4t'$.

We have to emphasize that the bipolaron moves already in the *first* order in polaron hopping. This remarkable property is entirely due to the strong on-site repulsion and long-range electron-phonon interaction that leads to a non-trivial connectivity of the lattice. This situation is unlike all other models studied previously. [Usually the bipolaron moves only in the second order in polaron hopping and therefore is very heavy.] In

our model, this fact combines with a weak renormalization of t' yielding a *superlight* bipolaron with mass $m^{**} \propto \exp(0.60\, E_p/\omega)$. We recall that in the Holstein model $m^{**} \propto \exp(2E_p/\omega)$. Thus the mass of the Fröhlich bipolaron scales approximately as the *cubic root* of that of the Holstein one.

At even stronger el-ph interaction, $V_c < 1.16E_p$, NNN bipolarons become stable. More importantly, holes can now form 3- and 4-particle clusters. Such clusters do not have resonant states and remain immobile in the first order in polaron hopping. The system quickly becomes a charge segregated insulator.

The superconductivity window that we have found, $1.16E_p < V_c < 1.23E_p$, is quite narrow. This indicates that the superconducting state in such systems is a subtle phenomenon which requires a fine balance between electronic and ionic interactions. Too strong el-ph interaction leads to clustering, while too weak interaction cannot bind the carriers and the superconductivity is at best of BCS type. These considerations may provide additional insight into the uniqueness of one particular structure, the copper-oxygen perovskite layer, to HTSC. It also follows from our model that superconductivity should be very sensitive to any external factor that affects the balance between V_c and E_p. For instance, pressure changes the octahedra geometry and hence E_p and $V_{\rm pa}$. Chemical doping enhances internal screening and consequently reduces E_p.

We now assume that the superconductivity condition is satisfied and show that our 'Fröhlich-Coulomb' model possesses many key properties of the underdoped cuprates. The bipolaron binding energy Δ should manifest itself as a normal state pseudogap with size of approximately half of Δ [6]. Such a pseudogap is indeed observed in many cuprates. There should be a strong isotope effect on the (bi)polaron mass because $t, t' \propto \exp(-\mathrm{const}\sqrt{M})$. Therefore the replacement of O^{16} by O^{18} increases the carrier mass [23]. Such an effect has been observed in the London penetration depth of the isotope-substituted samples [1]. The mass isotope exponent, $\alpha_m = d\ln m^{**}/d\ln M$, was found to be as large as $\alpha_m = 0.8$ in $La_{1.895}Sr_{0.105}CuO_4$. Our theoretical exponent is $\alpha_m = 0.3E_p/\omega$, so that the bipolaron mass enhancement factor is $\exp(0.6E_p/\omega) \simeq 5$ in this material. With the bare hopping integral $T_{NNN} = 0.2$ eV we obtain the in-plane bipolaron mass $m^{**} \simeq 10m_e$. Calculated with this value the in-plane London penetration depth, $\lambda_{ab} = [m^{**}/8\pi ne^2]^{1/2} \simeq 316$ nm (n the hole density) agrees well with the measured one $\lambda_{ab} \simeq 320$ nm. Taking into account the c-axis tunneling of bipolarons, the critical temperature of their Bose-Einstein condensation can be expressed in terms of the experimentally measured in-plane and c-axis penetration depths, and the in-plane Hall constant R_H as $T_c = 1.64f \cdot (eR_H/\lambda_{ab}^4\lambda_c^2)^{1/3}$. Here $f \approx 1$ and T_c, eR_H, and λ are measured in K, cm^3 and cm, respectively [24]. Using the experimental $\lambda_{ab} = 320$ nm, $\lambda_c = 4160$ nm, and $R_H = 4 \times 10^{-3}$ cm^3/C (just above T_c) one obtains $T_c = 31$ K in striking agreement with the experimental value $T_c = 30$ K. The recent observation of the normal state diamagnetism in $La_{2-x}Sr_xCuO_4$ [25] also confirms the prediction of the bipolaron theory [26]. Many other features of the bipolaronic (super)conductor, e.g., the unusual upper critical field, electronic specific heat, optical and tunneling spectra match those of the cuprates (for a recent review, see Ref. [27]).

In conclusion, we have studied a model with strong long-range electron-phonon and Coulomb interactions. The model shows a reach phase diagram depending on the ratio of the inter-site Coulomb repulsion and the polaronic (Franc-Condon) level shift. The ground state is a polaronic Fermi (or Luttinger) liquid at large Coulomb repulsions, a bipolaronic high-temperature superconductor at intermediate Coulomb repulsions, and a charge-segregated insulator at weak repulsion. In the superconducting phase, inter-site bipolarons are remarkably light leading to a high critical temperature. The model describes many properties of the superconducting cuprates.

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