Phonon-induced hole-hole effective interactions in the cuprates

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We have studied the phonon-induced effective interactions between charge carriers (holes) in a three-band model of the CuO_2 plane of high-temperature superconductors. For the phononic part we have included two oxygen phonon modes, breathing and buckling, and two types of electron-phonon interactions: namely, the ionic coupling, for which the displaced oxygens influence the energy level on copper, and the covalent coupling, for which the Cu-O hopping is modified by the displaced oxygens. Using a cell-perturbation method we have calculated the effective interaction between holes, V_{hh}, as induced by the electron-phonon couplings. We have found that the attractive interaction between holes, due to the breathing mode, is maximum in the presence of finite electron correlation. Furthermore, the presence of the buckling mode enhances the attractive interactions. We show that the calculated values of V_{hh} are of the same order as the values of the electronic pseudogap Δ, found in the normal state of the cuprates. We demonstrate that the isotope effect, related to the substitution $^{16}O \rightarrow ^{18}O$, changes the calculated values of V_{hh} in a way consistent with the experimentally found isotope effect of Δ. Finally, we have evaluated the superconducting transition temperature T_c and found a qualitative agreement with the experimental phase diagrams of the cuprates.

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I. INTRODUCTION

Despite great theoretical and experimental effort,[1] the nature of the pairing mechanism in high-temperature superconductors is still not understood. In principle, there are two possible mechanisms: electronic and phononic. The first one would result from strong correlation on copper ions,[2,3] whereas the second mechanism would be due to large electron-phonon (e-ph) coupling in these compounds.[4,5] Pure electronic models capture many well-established features of the cuprates, like antiferromagnetic order,[6] charge fluctuations and the stripe phase formation,[7] and anisotropy and symmetry of the superconducting state.[8] However, other phenomena, like the site-selective isotope effect,[9] frequency anomalies in neutron and Raman spectroscopy,[10,11] or tunneling measurements,[12] point directly to the importance of the e-ph interaction. They also clearly demonstrate the significance of particular in-plane, oxygen-related oscillations.[13]

Therefore, it seems natural to assume that both crucial factors, i.e., electron correlation and strong e-ph interaction, are present and that both play an important role. Particularly, in presence of a strong e-ph coupling, strong electron correlations may lead to new effects, absent in the standard BCS theory. Here, one should mention the anomalous behavior of the isotope effect, which strongly depends on doping.[14,15] Also, it was established that electron correlations enhance phonon-induced superconductivity[16] and are responsible for the d-wave symmetry of a superconducting order parameter.[17,18] Detailed studies of effective models with the e-ph coupling revealed the importance of the oxygen buckling mode in the pairing mechanism.[18,19] Using a more realistic three-band model of the CuO_2 plane, we showed that the oxygen breathing mode would also generate an attractive interaction between holes, provided the covalent e-ph coupling is included.[20–23]

One of the key features of high-temperature superconductors is the existence of a pseudogap in the electronic excitation spectra above the superconducting transition temperature T_c, i.e., in the normal state. The appearence of gaplike phenomena at a characteristic temperature $T^* > T_c$ has been found in a variety of compounds and by different methods.[24,25] Then, various possible mechanisms have been proposed in order to explain the origin of the pseudogap.[26] A huge isotope effect associated with the exchange of ^{16}O by ^{18}O (Ref. 27) undoubtedly points to the importance of the electron-lattice interaction in this phenomenon.

In this paper we consider the influence of electron correlation on the effective interaction between holes in a CuO_2 plane, as induced by both the breathing and buckling modes. As a method, we employ the cell-perturbation approach, used previously with success in studying electronic properties of the cuprates[28] as well as the bismuthates.[29] This paper is organized as follows. The model Hamiltonian and the cell-perturbation method are introduced in Sec. II. In Sec. III, we present the obtained numerical values of the effective interaction V_{hh}. Namely, we study (i) the effect of strong electron correlation on V_{hh} and (ii) the isotope substitution $^{16}O \rightarrow ^{18}O$ effect on V_{hh}, and we show (iii) how the calculated values of V_{hh} may change the values of T_c (superconducting transition temperature). Finally, Sec. IV gives the summary and conclusions of the paper.

II. MODEL HAMILTONIAN AND CELL-PERTURBATION METHOD

We consider a three-band model of the CuO_2 planes, which includes explicitly $d_{x^2-y^2}$ orbitals on copper and $p_{x/y}$ (p_σ) orbitals on oxygen,[21]

$$H=\sum_{i,l}\ \varepsilon_d[u_l,z_l]n_i^d+\sum_j\ \varepsilon_p n_j^p+\sum_i\ U_d n_{i,\uparrow}^d n_{i,\downarrow}^d$$
$$+\frac{m}{2}\sum_j\ (\dot{u}_j^2+\omega_b^2 u_j^2+\dot{z}_j^2+\omega_a^2 z_j^2)$$
$$+\sum_{i,l,\sigma}\ (-1)^{M_l}t_{pd}[u_l](d_{i,\sigma}^\dagger p_{l,\sigma}+\text{H.c.})$$
$$+\sum_{j,k,\sigma}\ (-1)^{N_{jk}}t_{pp}(p_{j,\sigma}^\dagger p_{k,\sigma}+\text{H.c.}), \qquad (1)$$

where i runs over all Cu sites $\mathbf{r}_i$ of a two-dimensional tetragonal lattice with primitive vectors $\mathbf{a}$ and $\mathbf{b}$, j and k run over all oxygen sites located (for given i) at $\mathbf{r}_i+\mathbf{a}/2$ and $\mathbf{r}_i+\mathbf{b}/2$, and the sum over l is for the four oxygen sites around $\mathbf{r}_i$, i.e., at positions $\mathbf{r}_i\pm\mathbf{a}/2$ and $\mathbf{r}_i\pm\mathbf{b}/2$. Operators $d_{i,\sigma}^\dagger$ and $p_{j,\sigma}^\dagger$ ($d_{i,\sigma}$ and $p_{j,\sigma}$) create (annihilate) holes in $3d_{x^2-y^2}$ orbitals on Cu and p_σ orbitals on O, respectively. The parameters ε_d and ε_p are the on-site energies on Cu and O, t_{pd} and t_{pp} are nearest-neighbor (NN) Cu-O and O-O hopping integrals, and U_d is the on-site Coulomb repulsion energy on copper. The numbers M_l and $N_{jk}=0$ or 1, according to the used phase convention.

The Hamiltonian (1) includes two types of oxygen phonon modes, namely, the breathing and buckling modes, with frequencies ω_b and ω_a, respectively; m is the oxygen ionic mass. Consistently with the phonon modes chosen, there are two types of e-ph coupling. First, the copper on-site energy is modulated by the oxygen displacements from their equilibrium positions: along the Cu-O bond (displacement u_l, e-ph strength λ_d) and perpendicularly to the plane (displacement z_l, e-ph strength λ_a). These interactions are diagonal in the sense that the oxygen coordinates are coupled to the total occupation number at copper, $n_i^d=n_{i,\uparrow}^d+n_{i,\downarrow}^d$. Second, we also consider the intersite coupling, in which the NN Cu-O hopping is modulated by the displacemets u_l in such a way that a longer (shorter) Cu-O bond results in a smaller (larger) Cu-O hybridization; here the coupling strength is denoted by λ_{dp}. Therefore, we write

$$\varepsilon_d[u_l,z_l]=\varepsilon_d^0+(-1)^{S_l}\lambda_d u_l+\lambda_a z_l, \qquad (2)$$

$$t_{pd}[u_l]=t_{pd}^0-(-1)^{S_l}\lambda_{dp}u_l, \qquad (3)$$

where the bare parameters are denoted as ε_d^0 and t_{pd}^0, and S_l ($=0$ or 1) is an appropriate phase factor.[21]

Here we would like to comment on two terms, which are not present in the starting Hamiltonian (1). First, in a more refined treatment, the on-site Coulomb repulsion energy on the oxygen ions, denoted as U_p, would be included.[30] Since U_p is much smaller than U_d and, moreover, we are mostly interested in the small doping region, then the effect of U_p may be safely neglected in our approach. The second absent term is connected with the so-called apical oxygens,[31] which play an important role in determining some properties of the cuprates. Including apical oxygen phonon modes creates some numerical problems, related to the bigger size of the Hilbert space. Even more important is that an accurate treatment should include anharmonicity, which is undoubtly present in the apical oxygen phonon modes.[32] Therefore, in this paper we restrict ourselves to the in-plane electronic and phononic degrees of freedom.

In order to use a cell-perturbation method, we perform canonical transformations for all oxygen-related operators (both electronic and phononic), thus transforming the Hamiltonian (1) into a new representation in which all operators are centered on copper sites. After the transformation to a new basis is done, the Hamiltonian is split into two parts: a diagonal part, which includes only operators related to a single copper site, and terms representing interaction between sites:

$$H=H_0+H_1=\sum_i\ h_{0i}+\sum_{ij}\ h_{ij}, \qquad (4)$$

where h_{0i} is the Hamiltonian of the ith cell and $h_{ij}=T_{ij}+V_{ij}$ is a part describing interactions between cells; T_{ij} are hoppings between cells and V_{ij} are intercell static interactions. The details of this procedure together with the expressions for h_{0i}, T_{ij}, and V_{ij} are presented elsewhere.[21,28] Here we would like to stress that with this choice of reference system, H_0 contains already a big fraction of the many-body correlation and e-ph effects, all treated on equal footing. As a next step, a single-cell Hamiltonian h_{0i} is solved, using the exact diagonalization technique and which yields the eigenvalues e_ν and eigenvectors $|\nu,i\rangle$, with ν representing all quantum numbers (electronic and phononic). For simplicity, we limit ourselves only to three lowest-energy cases: the zero-hole state $|0,i\rangle$, the one-hole state $|g,i\rangle$, and the two-hole singlet state $|S,i\rangle$, as other states are much higher in energy; the two-hole singlet state is usually refered to as the Zhang-Rice (ZR) singlet.[33] Then the Hamiltonian is expressed in the cell basis by means of the Hubbard X operators:[34]

$$H=\sum_i\ \sum_\nu\ e_\nu X_i^{\nu\nu}+\sum_{ij}\ \sum_{\alpha\beta\gamma\delta}(t_{ij}^{\alpha\beta\gamma\delta}+v_{ij}^{\alpha\beta\gamma\delta})X_i^{\alpha\gamma}X_j^{\beta\delta}, \qquad (5)$$

where $X_i^{\alpha\gamma}=|\alpha,i\rangle\langle\gamma,i|$. The effective Hamiltonian parameters

$$t_{ij}^{\alpha\beta\gamma\delta}=\langle\alpha,i|\langle\beta,j|T_{ij}|\gamma,i\rangle|\delta,j\rangle, \qquad (6a)$$

$$v_{ij}^{\alpha\beta\gamma\delta}=\langle\alpha,i|\langle\beta,j|V_{ij}|\gamma,i\rangle|\delta,j\rangle \qquad (6b)$$

are the hopping amplitudes and static interactions between cells, respectively. A similar approach for transforming the Hamiltonian, the so-called standard-basis operator method, was developed in Ref. 35. Out of all the matrix elements representing static interactions (6b), the most important are the diagonal ones between the NN sites,[36] i.e., $v^{\alpha\beta}=\langle\alpha,i|\langle\beta,j|V_{ij}|\alpha,i\rangle|\beta,j\rangle$, where $\alpha,\beta=g$ or S; here i and j denote NN sites.

In the insulating case there is, on average, one hole per a CuO_2 cell, so the energy of hole-hole interaction is Nv^{gg}, where N is the number of all NN pairs of sites in the plane. In a doped material, there are additionaly interactions be-

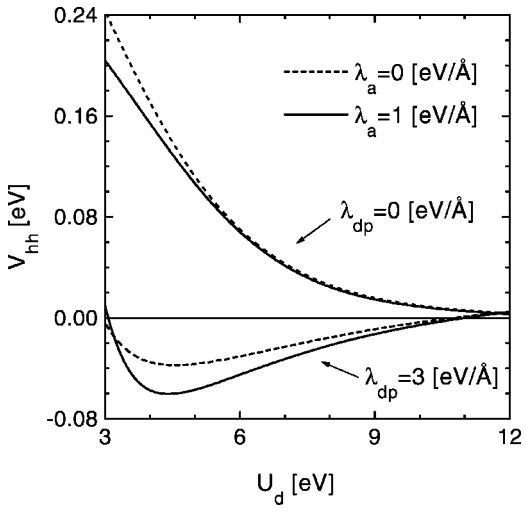

FIG. 1. Effective hole-hole interaction between the charge carriers, V_{hh}, as a function of the on-site copper correlation energy U_d and for different values of the electron-phonon couplings λ_{dp} and λ_a. All the curves are for $\lambda_d=3$ Å/eV.

tween one-hole states and ZR-singlet states, v^{Sg}, and between two ZR-singlet states, v^{SS}. Having calculated all these interactions, we can evaluate the effective interaction between two doped holes into a CuO_2 plane. We assume a small doping of the system, in which overlap between hole pairs is negligible. When two doped holes are far apart, each of them interacts with four holes around with energy v^{Sg}, so the total energy of interaction in the system is

$$E_\infty = 8v^{Sg} + (N-8)v^{gg}. \tag{7}$$

When they are on NN sites, they interact with three one-hole states and with each other through v^{SS}, and there is an extra one-hole bond in the plane, so the energy is

$$E_{NN} = v^{SS} + 6v^{Sg} + (N-7)v^{gg}. \tag{8}$$

The effective net interaction between two doped holes can be calculated as the difference in energy in these two cases:

$$V_{hh} = E_{NN} - E_\infty = v^{SS} + v^{gg} - 2v^{Sg}. \tag{9}$$

III. EFFECTIVE INTERACTION BETWEEN CHARGE CARRIERS

We study the dependence of the effective hole-hole interaction on electron correlation, U_d. As in our previous works,[20–23] we have fixed the values of the parameters (except U_d), according to the so-called standard set.[30] Namely, we adopt the following values: $\epsilon_p - \epsilon_d[0,0] = 3.5$, $t_{pd}[0] = 1.3$, and $t_{pp} = 0.65$ (all in eV). We have also fixed the values of one diagonal *e*-ph coupling $\lambda_d = 3$ eV/Å (Ref. 37) and the phonon frequencies $\hbar\omega_a = 40$ meV, $\hbar\omega_b = 70$ meV.[10–12]

A. Role of electronic correlation

In Fig. 1 we plot V_{hh} as a function of U_d, for different values of λ_{dp} and λ_a. For $\lambda_{dp}=0$, the interaction is always repulsive and decreases with increasing U_d. With increasing λ_{dp}, the possibility of an attractive interaction arises. For fixed, nonzero λ_{dp}, there is an interval of U_d for which V_{hh} becomes attractive. For stronger λ_{dp}, that interval gets wider and the attraction becomes stronger. The overall conclusion is that for the effective interaction between the charge carriers to become attractive, the optimum is a moderate value of correlation, though for larger values of the *e*-ph coupling, this optimum value of correlation gets smaller. The physical reason for this is that for small values of U_d, the interaction is repulsive because holes are localized on copper ions and the effect of the covalent coupling is much reduced. When correlations are very strong, holes are localized on oxygens, and the effect of Cu-O hopping is again considerably suppressed. The presented results demonstrate that for the parameters chosen, the most favorable conditions for superconductivity are when correlations are neither too weak nor too strong.

For the high-T_c cuprates, the actual value of U_d is around 8 eV (Ref. 30) and such a value of the correlation should be considered as not giving the maximum phonon-induced attraction between the charge carriers. Therefore, we do not claim that, from the superconductivity point of view, the correlations in the cuprates are optimal. But there is no doubt that the obtained values of V_{hh} are the attractive ones and that the phonon-induced effective interaction V_{hh} supports superconductivity. Second, a more accurate determination of the *e*-ph interaction couplings involved, particularly λ_a, might give higher values and thus lead to even more favorable conditions for superconductivity to occur. The results presented in Fig. 1 also demonstrate that the stronger the *e*-ph coupling, the greater the optimum value of U_d. This is also demonstrated when the effect of the buckling mode is studied by putting $\lambda_a \neq 0$. The overal picture is similar, though we see that the diagonal *e*-ph coupling, related to the buckling mode, stabilizes the attractive interactions furthermore.

B. Isotope effect of V_{hh}

We have also examined how the oxygen ion substitution $^{16}O \rightarrow {}^{18}O$ changes the values of the calculated effective interactions. For simplicity, we have assumed that the phonon frequencies scale like $\sim\sqrt{1/m}$ [see Eq. (1)]. This simplification seems reasonable, as the copper ions are much heavier and the ionic effective mass should be very close to the mass of oxygen. We have repeated all the calculations but with scaled values of the phonon frequencies ω_a and ω_b. Figure 2 presents the obtained values of V_{hh} as a function of U_d. The inset of Fig. 2 shows the calculated difference $\Delta V_{hh} = V_{hh}(^{18}O) - V_{hh}(^{16}O)$ for two considered values of the oxygen ionic mass. These values should be considered as the isotope effect results for V_{hh}. The isotope effect is negative for moderate values of the electron correlation, approximately for U_d in the interval between 4 and 8 eV. For very small and very large correlations, the isotope effect of V_{hh} is positive.

Now, we would like to discuss the obtained results and their connection with the experimental results for the

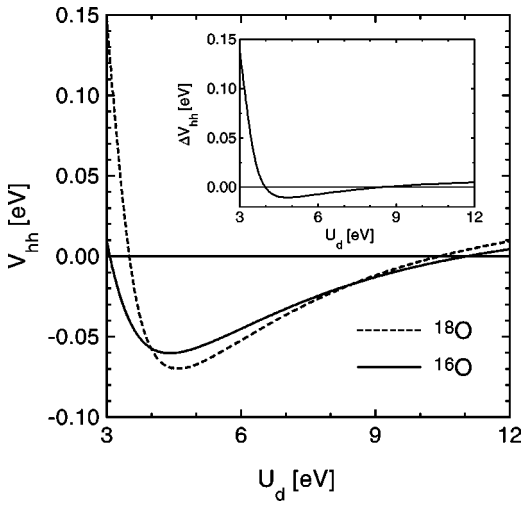

FIG. 2. Effective hole-hole interactions V_{hh} between the charge carriers as functions of the on-site copper correlation energy U_d and for two values of the oxygen ion mass. Both the curves are for $\lambda_d = 3$ eV/Å, $\lambda_{dp} = 3$ eV/Å, and $\lambda_a = 1$ eV/Å. In the inset, the isotope effect values $\Delta V_{hh} = V_{hh}(^{16}\mathrm{O}) - V_{hh}(^{18}\mathrm{O})$ are shown.

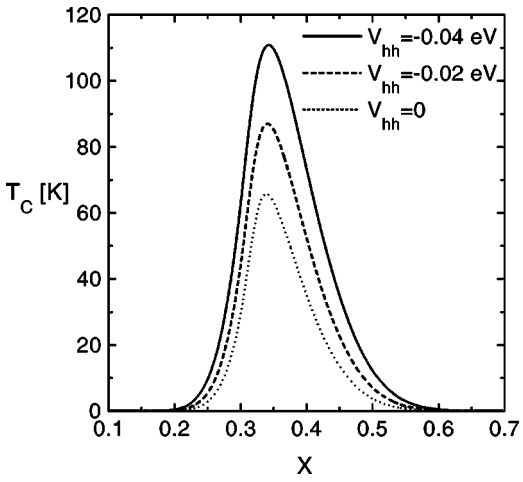

FIG. 3. Superconducting transition temperature T_c as a function of hole concentration x for three different values of the phonon-induced effective interaction V_{hh}. Only d-wave symmetry results are presented.

pseudogap Δ. The typical observed values of the pseudogap Δ are between 10 and 60 meV.[27] Looking at Figs. 1 and 2 we see that the obtained values of V_{hh} are exactly of the same order of magnitude. It is remarkable that such a simple model as considered in this paper could give a hole-hole interaction of the same order as Δ. It is physically reasonable to expect that the hole-hole interaction V_{hh} would be of the same order as Δ, although to calculate Δ, values of the effective NN hopping should be known. Finding the values of the effective hopping is a highly nontrivial task and is beyond the scope of the present paper.[21] Also, our isotope effect results for V_{hh} (Fig. 2) agree qualitatively with the experimental data.[27]

The obtained values of V_{hh} and ΔV_{hh} should be considered only as the qualitative ones, as the exact values of the model parameters, mainly U_d and all the e-ph couplings, are still unknown to a better precision. However, we have demonstrated clearly that the considered model captures the essential physics of the CuO_2 plane, particularly with respect to the generation of the effective interactions between the charge carriers.

C. Superconducting transition temperature

Finally, we have also evaluated the values of the superconducting transition temperature T_c, using the mean-field Hubbard X-operator technique,[34,38] which proved useful for studying properties of strongly correlated systems. To calculate T_c, we adopt the approximate one-band t-t'-J model, which includes the effective hoppings (t,t') and antiferromagnetic interaction (J). This model is completed by the phonon-derived static interaction V_{hh}, which within the X-operator technique is added to J, thus forming the total pairing interaction. We have fixed $J = 0.2$ eV,[28] which is of the order of the experimental value. To complete the set of parameters, the effective hopping between different sites should be specified. Following their work on the one-band effective Hamiltonian,[28] Feiner *et al.*[39] argued that the effective hoppings of different ranges (t for NN, t' for next NN, t'' for next-next NN) should be a crucial factor that differentiates between the cuprates. However, for qualitative estimates, we adopt some average values, namely, $t = 0.4$ eV, $t' = -0.06$ eV, and $t'' = 0$.[30,39] It is well known that the e-ph coupling leads to retardation effects, known also as the band narrowing. This effect may be quite strong, particularly if the average over the ground-state phononic wave function is made.[40] In the cell-perturbation method, a large portion of the e-ph coupling effects is already in the zeroth-order terms. This is particularly true for the copper-oxygen hopping terms, i.e., those proportional to t_{pd} [see Eq. (1)]. That is the first reason why we expect the retardation effects to be much reduced in our approach. Moreover, as we demonstrated for cubic bismuth superconductors,[29] retardation effects should be much reduced if a more accurate phonon wave function is used, for example, from a finite-cluster evaluation. Therefore, for the qualitative predictions we are making, we assume that the effective hopping integrals t and t' have fixed values, namely, those derived by the cell-perturbation method, as applied for the electronic Hamiltonian alone.[39] We admit that using a more accurate phononic wave function would produce some band narrowing, which eventually might lead to higher values of T_c. With this in mind, the obtained values of T_c should be considered as a lower bound that follows from the considered model.

Figure 3 presents the calculated values of T_c of the d-wave symmetry, as relevant for the cuprates. Our model essentially reproduces the correct shape of the experimental phase diagram of the cuprates. There is a finite region with nonzero transition temperature, and T_c at optimum doping has the correct value around 100 K. Another interesting feature is a very high sensivity of T_c to small variations of V_{hh}. By changing V_{hh} from 0 (no phonon-induced interaction) to -0.02 eV (physical value for the cuprates, as seen in Fig. 1), T_c increases from about 60 to about 90 K. That means that

even though the electronic antiferromagnetic interaction J is not sufficient to give T_c high enough, a small admixture of the phonon-induced interaction V_{hh} makes a high superconducting transition temperature possible. We believe that cooperation of the electronic and phononic mechanism in the spirit just described might be a driving force of high-temperature superconductivity.

Here we do not study the direct isotope effect, related to T_c. We think that such a study would require a more sophisticated treatment. For example, one should take into account variation of both J and V_{hh} with hole concentration, the effect which we neglect in this paper entirely. Such a treatment, which would be a next step to the first approximation we make here, with concentration-dependent parameters, might give slightly different values of T_c, especially for higher doping. However, we think that the general overall picture would survive and the obtained phase diagram should have a similar shape.

IV. CONCLUDING REMARKS

In this paper, we have studied the effective interactions between the charge carriers in the CuO_2 planes of high-temperature superconductors. Particularly, we considered a three-band extended Hubbard model together with two important in-plane oxygen-related phonon modes: breathing and buckling. Additionally, to previously found results about the importance of these phonon modes, here we demonstrate that in the presence of the electron-phonon interaction, the electron correlation should be neither too small nor too big, if attractive effective interactions between the charge carriers are to be expected. In that sense, for fixed values of the electron-phonon couplings, there exists an optimum value of the electron correlation, for which the attractive interaction gets a maximum. This result gives important limitations to existing models of high-temperature superconductors. We have also demonstrated that the obtained values of the effective interaction and its change with oxygen mass (isotope effect) are consistent with the measured pseudogap values. Finally, we have demonstrated that the total effective interaction, built up of the antiferromagnetic interaction J and the phonon-induced interaction V_{hh}, would give a qualitatively and semiquantitatively correct phase diagram of the cuprates.

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