Electron-phonon interaction in the cuprates: Breathing versus buckling mode

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Using a cell perturbation method, we investigate the role played by two important phonon modes in high-temperature superconductors, namely, the in-plane-oxygen breathing and buckling modes. We demonstrate that diagonal electron-phonon interactions, present in both modes, lead to opposite interactions: attraction for the buckling and repulsion for the breathing mode. When the nondiagonal electron-phonon interaction is included with the breathing mode, the possibility of attraction also arises within this mode. We also show that the effective Hamiltonian may be reduced to a Hubbard-like model with a simple, renormalized electron-phonon interaction. [S0163-1829(99)14121-3]

There are three distinctive features of high-temperature superconducting cuprates[1]: strong correlations on copper,[2] pronounced anisotropy,[3,4] and large electron-phonon interactions.[5] In the simplest case, correlations may be accounted for by a single-band Hubbard model.[6] However, under hole doping, the extra holes are mainly of oxygen p_σ character, and this has led to a debate as to whether an effective single-band model could adequately describe the essential physical properties of the cuprates. Emery[7] and Varma, Schmitt-Rink, and Abrahams[8] proposed a three-band (d-p) model that includes $3d_{x^2-y^2}$ and $2p_\sigma$ orbitals explicitly. Subsequently, Zhang and Rice (ZR)[9] argued that an extra hole, introduced by doping, is shared between four oxygen atoms and couples strongly to a central Cu spin, thus forming a local singlet. In this framework, the motion of holes is equivalent to the hopping of singlets between two nearest-neighbor (NN) copper sites. Further work on the reduction of d-p models to an effective single-band model for low-energy physics has provided convincing support for this physically appealing picture.[10,11]

The main physics of the cuprates takes place in the CuO_2 planes, with relatively weak interplanar coupling.[3] Therefore, to a first approximation, one may start by considering a single CuO_2 plane, as we do in this paper. However, in a more accurate treatment, the so-called apical oxygens should be also taken into account.[12–14]

There is now general agreement that the electron-phonon (e-ph) interaction in the cuprates is large, though the importance of its role, in competition with strong correlations and anisotropy, remains uncertain and controversial, particularly its possible contribution to superconductivity. Here we just mention that theoretical, self-consistent, band-structure calculations give a large e-ph coupling of $\lambda \sim 1-1.5$,[5] and that experimental results suggest a substantial e-ph coupling in the cuprates.[15] In particular, the oxygen isotope effect has been observed in both underdoped and overdoped systems,[16] and site-selective measurements[17] show that the dominant contribution ($>80\%$) is from in-plane oxygen. The polaronic nature of doped charge carriers in the cuprates, observed in various optical experiments,[18] is further evidence of strong e-ph coupling in these materials. Tunneling experiments, mainly of Bi2212 systems, also directly show the phonon contribution to high-T_C superconductivity,[19] and detailed comparison of the spectral-function of the e-ph interaction $\alpha^2F(\omega)$ with the phonon spectrum of Bi2212 reveals that the low-frequency vibrations contribute mostly to the e-ph coupling.[20] These are the axial (buckling) oxygen modes and cation vibrations (Cu, Ca, and Sr). There is also a considerable contribution to $\alpha^2F(\omega)$ from the high-frequency oxygen breathing modes.

In this paper we study a single CuO_2 plane, described by a three-band model, together with two important phonon modes. They are related to the oxygen vibrations, and these are the breathing and buckling modes. We consider three electron-phonon interactions, two (diagonal and nondiagonal) associated with breathing and one (diagonal) associated with the buckling mode. Using the cell perturbation method, which proved useful in deriving the effective Hamiltonians for electronic systems,[10,11] we study the effects of these couplings on the effective interactions between the charge carriers in the CuO_2 plane. We also derive the form of an effective Hubbard-like Hamiltonian, in the presence of the electron-phonon interactions considered.

We start from the following three-band Hamiltonian of a single CuO_2 plane with the e-ph couplings arising from in-plane and axial motions of oxygen ions,

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

where i runs over all Cu sites $\mathbf{r}_i$ of a tetragonal lattice with primitive vectors $\mathbf{a}$ and $\mathbf{b}$, j and k run over all oxygen sites, and the sum over l enumerates the four oxygen sites around $\mathbf{r}_i$ at positions $\mathbf{r}_i \pm \mathbf{a}/2$ and $\mathbf{r}_i \pm \mathbf{b}/2$. Operators $d^{\dagger}_{i,\sigma}$ and $p^{\dagger}_{j,\sigma}$ create holes in $3d_{x^2-y^2}$ orbitals on Cu and p_σ orbitals on O, with number operators $n^d_{i,\sigma}=d^{\dagger}_{i,\sigma}d_{i,\sigma}$ and $n^p_{j,\sigma}=p^{\dagger}_{j,\sigma}p_{j,\sigma}$ for spin $\sigma=\uparrow$ or $\downarrow$. We use a phase convention such that the orbitals p_σ transform like x or y, and hence the numbers M_l and $N_{jk}=0$ or 1, according to this convention. The parameters ε_d and ε_p are on-site energies on Cu and O, while t_{pd} and t_{pp} are NN Cu-O and O-O hopping integrals, respectively, and U_d is the on-site Coulomb repulsion energy on copper.

Hamiltonian (1) includes two types of oxygen vibrations, i.e., breathing and buckling modes, with frequencies ω_b and ω_a, respectively, and m is the mass of oxygen. The e-ph interactions considered are linear in atomic displacements. In the first term, the copper on-site energy is modulated by the oxygen displacements from their equlibrium positions: in-plane u_l (breathing mode, e-ph constant λ_d)[21,22] and axial z_l (buckling mode, e-ph constant λ_a).[22–24] Both these interactions are diagonal in the sense that the displacements are coupled to the total occupation number at copper, $n^d_i=n^d_{i,\uparrow}+n^d_{i,\downarrow}$. In the coupling to the breathing mode, the energy increases with Cu-O bond length, and hence $S_l=0$ for $\mathbf{r}_l=\mathbf{r}_i+\mathbf{e}$, and $S_l=1$ for $\mathbf{r}_l=\mathbf{r}_i-\mathbf{e}$, where $\mathbf{e}=\mathbf{a}/2$ or $\mathbf{b}/2$. To be consistent, we also consider the intersite, covalentlike coupling, in which the NN, Cu-O hopping is modulated by the displacemets u_l, in such a way that longer (shorter) Cu-O bond gives smaller (larger) Cu-O hybridization, the coupling constant in this interaction is denoted by λ_{dp}.[21]

A few further comments justifying the form of the e-ph interaction are in order. First, the diagonal e-ph interactions result from changes of the copper ionic potential when all four NN oxygen ions move simultaneously. For both modes considered, there is no linear change of the crystal potential at the oxygen sites since these ions move about their equilibrium positions. Hence there is no direct e-ph coupling between doped holes on oxygen to first order in the oxygen coordinates x_l and z_l. This contrasts with the model of an isolated CuO_2 molecule considered in Ref. 13, relevant to vibrations of CuO chains in the $YBa_2Cu_3O_7$ compounds. Second, there is an indirect effect of the oxygen hole doping on the copper ionic potential and thus on the e-ph interaction. With doping on oxygen, the effective e-ph couplings λ_a and λ_d will thus change. However, this change is expected to be small and we neglect it, except in an average way by considering different values of the e-ph couplings. We also limit ourselves to oxygen vibrations only, since the oxygen ions are much lighter than the copper. Finally, the key difference between the considered breathing and buckling modes lies in the phase factor $(-1)^{S_l}$ [see Eq. (1)]. As we show below, both modes give rise to different effective interactions between charge carriers.

We introduce the boson representation for the phononic part of Eq. (1) by substitutions

$$u_l=\sqrt{\frac{\hbar}{2m\omega_b}}(e^+_l+e_l), \overset{def}{=} \xi_d(e^+_l e_l), \tag{2a}$$

$$z_l=\sqrt{\frac{\hbar}{2m\omega_a}}(f^+_l+f_l) \overset{def}{=} \xi_a(f^+_l+f_l), \tag{2b}$$

where e and f are the bosonic operators. We apply a cell-perturbation method, which consists of the following. First, we transform all operators relating to oxygen sites, i.e., fermionic $p_{l,\sigma}$ and bosonic e_l and f_l operators, to a basis centered on copper. For the fermionic oxygen orbitals, we construct orthogonal Wannier functions $a_{i,\sigma}$ and $b_{i,\sigma}$ by transforming the original orbitals to the reciprocal $\mathbf{q}$ space, then introducing ''canonical fermions''[25,10]

$$a_{\mathbf{q},\sigma}=-\frac{i}{\mu_{\mathbf{q}}}(s_{\mathbf{q},y}p_{\mathbf{q},x,\sigma}+s_{\mathbf{q},x}p_{\mathbf{q},y,\sigma}), \tag{3a}$$

$$b_{\mathbf{q},\sigma}=\frac{i}{\mu_{\mathbf{q}}}(s_{\mathbf{q},x}p_{\mathbf{q},x,\sigma}-s_{\mathbf{q},y}p_{\mathbf{q},y,\sigma}), \tag{3b}$$

where $s_{\mathbf{q},x}=\sin(q_x/2)$, $s_{\mathbf{q},y}=\sin(q_y/2)$, and $\mu_{\mathbf{q}}=\sqrt{s^2_{\mathbf{q},x}+s^2_{\mathbf{q},y}}$. Transforming back to real space yields $a_{i,\sigma}$ and $b_{i,\sigma}$. Similarly, phononic operators are introduced by the canonical transformations

$$A_{\mathbf{q}}=-\frac{i}{\mu_{\mathbf{q}}}(s_{\mathbf{q},x}e_{\mathbf{q},x}+s_{\mathbf{q},y}e_{\mathbf{q},y}), \tag{4a}$$

$$B_{\mathbf{q}}=-\frac{i}{\mu_{\mathbf{q}}}(s_{\mathbf{q},y}e_{\mathbf{q},x}-s_{\mathbf{q},x}e_{\mathbf{q},y}), \tag{4b}$$

$$C_{\mathbf{q}}=\frac{1}{\nu_{\mathbf{q}}}(c_{\mathbf{q},x}f_{\mathbf{q},x}+c_{\mathbf{q},y}f_{\mathbf{q},y}), \tag{4c}$$

$$D_{\mathbf{q}}=\frac{1}{\nu_{\mathbf{q}}}(c_{\mathbf{q},y}f_{\mathbf{q},x}-c_{\mathbf{q},x}f_{\mathbf{q},y}), \tag{4d}$$

where $c_{\mathbf{q},x}=\cos(q_x/2)$, $c_{\mathbf{q},y}=\cos(q_y/2)$, $\nu_{\mathbf{q}}=\sqrt{c^2_{\mathbf{q},x}+c^2_{\mathbf{q},y}}$, with $e_{\mathbf{q},x}$, $e_{\mathbf{q},y}$, $f_{\mathbf{q},x}$ and $f_{\mathbf{q},y}$ the Fourier transforms of the corresponding phononic operators. Transforming back into real space, the Hamiltonian may be expressed in the form

$$H=H_0+H_1, \tag{5}$$

where $H_0=\Sigma_i h_{i0}$ is the Hamiltonian for noninteracting cells, with h_{i0} the Hamiltonian of the ith cell and H_1 representing cell-cell interactions.[26] With this form of the Hamiltonian, much of the correlation and e-ph effects are already included in H_0. Moreover, both these effects are treated on an equal footing, unlike in the more usual adiabatic or antiadiabatic approximations.[27]

Before considering the effective interaction between the charge carriers (holes), the one-cell Hamiltonian h_{i0} has to be solved, either analytically or (more usually) numerically. The detailed procedure of obtaining the solution will be presented elsewhere.[26] Here we point out that it is sufficient to solve h_{i0} for zero, one, and two holes, as states with more holes are much higher in energy. For the underlying d-p model of the electron subsystem, we take the ''standard set'' of parameters, i.e., $t_{pd}=1.3$ eV, $t_{pp}=0.65$ eV, $\varepsilon_p-\varepsilon_d=3.5$ eV, and $U=U_d=9$ eV.[28] In numerical calculations, phonon energies are taken as $\hbar\omega_b=0.1$ eV and $\hbar\omega_a=0.05$ eV for the breathing and buckling modes, respectively. These cor-

respond closely to the measured frequencies.[20] The lowest-energy states which belong to different subspaces are denoted as $|0_k\rangle$, $|g_k\rangle$, and $|S_k\rangle$, being the generalized vacuum, doublet, and ZR singlet states, respectively. The index k labels the phonon-induced ladder of vibronic states, belonging to a particular electronic state.

We stress that in the cell-perturbation method, the one-cell solutions already contain not only the main correlation effects,[10,11] but also most of the effect of the strong e-ph interaction. We have investigated the behavior of the one- and two-hole solutions in detail. Detailed results of this investigation will be presented elsewhere,[29] with only the main qualitative findings given here. First, the polaronic effects are stronger for larger values of the diagonal e-ph interaction (λ_d and λ_a), while the off-diagonal e-ph coupling makes the one-cell solution less polaronic. This is consistent with the single-band single-phonon-mode model, where all the polaron effects are diagonal in origin.[30] Second, we have found a very weak phonon frequency renormalization ($<1\%$). Third, it appears that in a multiband, multiple-phonon-mode model, the polaronic effects are less pronounced than in a comparable single-band single-phonon-mode model.[29]

Generally, $H_1=T_1+V_1$, where T_1 and V_1 represent the hopping and the static interactions between cells, respectively. In this paper, we do not consider the hopping part of the Hamiltonian, but concentrate on the effect of e-ph couplings on static interactions between charges. These interactions result from the on-site e-ph coupling to both the breathing and axial modes,

$$V_1=H_{\rm br}+H_{\rm ax}=2\lambda_d\xi_d\sum_{ij}\ \mu_{ij}(A_i^\dagger+A_i)n_j^d$$

$$+2\lambda_a\xi_a\sum_{ij}\ \phi_{ij}(C_i^\dagger+C_i)n_j^d$$

$$\overset{def}{=}\sum_{ij}\ V_{ij}\,, \tag{6}$$

where $\mu_{ij}=1/N\Sigma_{\mathbf{q}}\mu_{\mathbf{q}}\exp[-i\mathbf{q}(\mathbf{r}_i-\mathbf{r}_j)]$ and $\phi_{ij}=1/N\Sigma_{\mathbf{q}}v_{\mathbf{q}}\exp[-i\mathbf{q}(\mathbf{r}_i-\mathbf{r}_j)]$ are the coefficients resulting from the canonical transformation [Eqs. (4a)–(4d)]. Furthermore, this term may be expressed in terms of Hubbard operators, $X_i^{\alpha\beta}=|\alpha,i\rangle\langle\beta,i|$, as follows:

$$V_1=\sum_{ij}\ \sum_{\alpha\beta\gamma\delta}\langle\alpha,i;\beta,j|V_{ij}|\gamma,i;\delta,j\rangle X_i^{\alpha\gamma}X_j^{\beta\delta}\,, \tag{7}$$

where V_{ij} is given by Eq. (6), with α, β, γ, and δ denoting all quantum numbers of the eigenstates. Equation (7) may be written in the simple form

$$V_1=\sum_{\langle ij\rangle}\ \sum_{\alpha\beta}\ [v_{\alpha\beta}X_i^{\alpha\alpha}X_j^{\beta\beta}+\Lambda_{\alpha\beta}X_j^{\beta\beta}(C_{i,\alpha}^\dagger+C_{i,\alpha})$$

$$+\Lambda_{\beta\alpha}X_i^{\alpha\alpha}(C_{j,\beta}^\dagger+C_{j,\beta})]\,, \tag{8}$$

where $v_{\alpha\beta}=\langle\alpha,i;\beta,j|V_{ij}|\alpha,i;\beta,j\rangle$ are the diagonal matrix elements ($\alpha,\beta=0$, g, or S); $\Lambda_{\alpha\beta}$ are effective e-ph interaction strengths, which are quite small (e.g., $\Lambda_{gg}=0.02$ eV, $\Lambda_{SS}=0.03$ eV); and $C_{i,\alpha}=\Sigma_k(\pm)\sqrt{k}X_i^{\alpha_{k-1}\alpha_k}$ are bosonic operators. Thus the static interaction takes a very simple form which would be precisely that of a single band if all $\Lambda_{\alpha\beta}$ were equal.[31] This is analogous to the derivation of the effective single-band model from a multiple-band model,[11] but with phonon modes included.

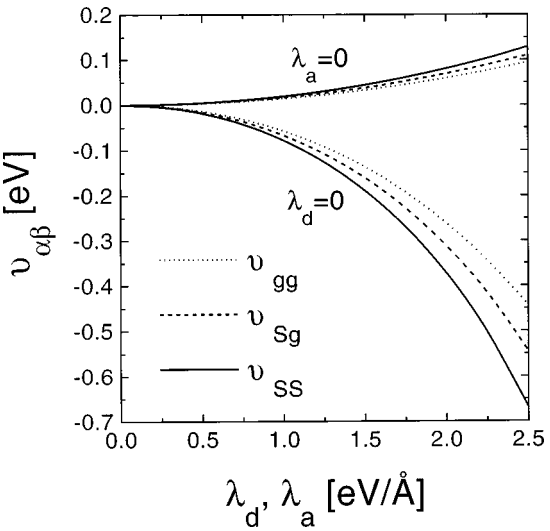

FIG. 1. Effective interactions v_{gg}, v_{Sg}, and and v_{SS} between the charge carriers, as functions of the electron-phonon couplings. All curves are for $\lambda_{dp}=0$. The upper three curves are the functions of λ_d with $\lambda_a=0$, while the lower ones are the functions of λ_a with for $\lambda_d=0$.

Let us examine particular terms in V_1, which arise from the e-ph coupling to the breathing and buckling modes. These terms $H_{\rm br}$ and $H_{\rm ax}$ have the same form but different coefficients, whose magnitudes depend on the phonon frequencies with sign determined by μ_{ij} and ϕ_{ij}, respectively. For NN interaction, we prove that

$$\mu_{01}=\frac{1}{N}\sum_{\mathbf{k}}\ \sqrt{\sin^2\frac{k_x}{2}+\sin^2\frac{k_y}{2}}\cos k_x$$

$$=-\frac{1}{N}\sum_{\mathbf{q}}\ \sqrt{\cos^2\frac{q_x}{2}+\cos^2\frac{q_y}{2}}\cos q_x$$

$$=-\phi_{01}\,. \tag{9}$$

Since the sign of the effective hole-hole interaction induced by the e-ph couplings is determined by the sign of coefficients μ_{01} or ϕ_{01}, the two phonon modes, breathing and buckling, generate interaction energies of opposite sign. From the numerical calculations we have shown that, for $\lambda_{dp}=0$, the breathing mode results in repulsive interaction between holes and the buckling mode in attractive interaction between holes. These findings are illustrated in Fig. 1, where the values of v_{gg}, v_{Sg}, and v_{SS} for NN cells are plotted in two different situations $\lambda_d\neq0$, $\lambda_a=0$ and $\lambda_d=0$, $\lambda_a\neq0$. Similarly, in Fig. 2 we plot the effective NN interaction between holes $V_{hh}=v_{gg}+v_{SS}-2v_{Sg}$ (Ref. 11) as functions of two e-ph couplings λ_d and λ_a. The magnitude of the attraction induced by the buckling mode is comparable to that obtained by Nazarenko and Dagotto,[23] and supports results of other investigations.[24] The case in which both modes contribute to the effective interactions will be checked numerically in a subsequent paper. In Fig. 3, we

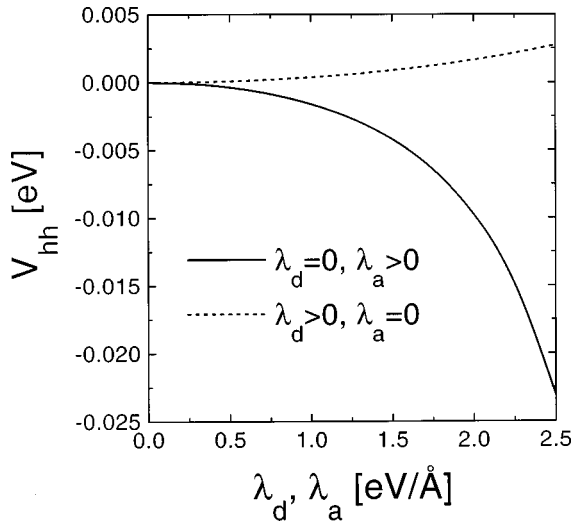

FIG. 2. Effective hole-hole interactions V_{hh}, between the charge carriers, as functions of the electron-phonon couplings. Both curves are for $\lambda_{dp}=0$. The upper curve is the function of λ_d with $\lambda_a=0$, while the lower one is the function of λ_a with $\lambda_d=0$.

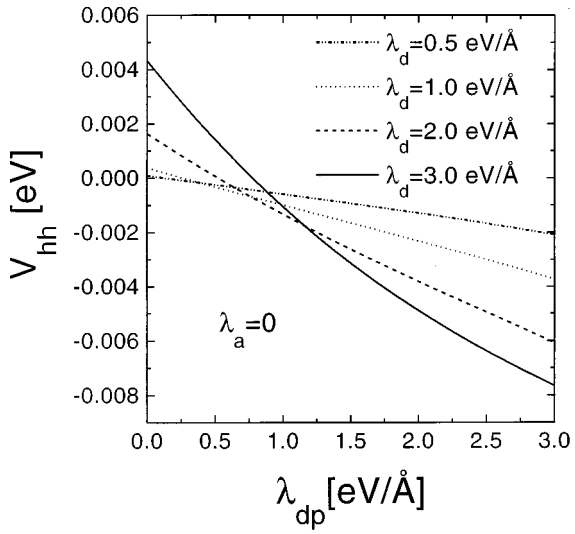

FIG. 3. The same as in Fig. 2, but as the functions of λ_{dp} for four values of λ_d; $\lambda_a=0$.

show the effect of the nondiagonal part of the *e*-ph interaction. We see that, for a given value of λ_d, the hole-hole interaction becomes attractive for large enough λ_{dp}, which enters the effective interaction only through the one-cell solution. The presence of the attractive interaction between the charge carriers, in the presence of the breathing mode with the non-diagonal *e*-ph interaction, is a result which in our opinion may strongly support the idea that the *e*-ph interaction is one of the key factors for understanding the high-temperature superconductivity. The contribution of both modes, breathing and buckling, to the pairing mechanism is in agreement with the tunneling spectroscopy, which reveals the important role of these phonon modes in the superconducting state.[19,20]

In this paper we have considered a Hamiltonian for a single CuO_2 plane with two phonon modes, breathing and buckling, and the associated electron-phonon interactions: diagonal for both modes and nondiagonal for the breathing mode. We have shown that the diagonal parts of the electron-phonon interactions have opposite effects for the effective interactions between the charge carriers: attraction for the buckling mode and repulsion for the breathing mode. However, when the nondiagonal interaction is included, it appears that also the breathing mode may lead to attraction. We have also presented a simple, effective one-band Hamiltonian for a CuO_2 plane, which takes the form of a single-band Hubbard model with small electron-phonon interactions.

Since the model we have considered is restricted to a CuO_2 plane, only phononic modes resulting from the movement of the in-plane oxygens are taken into account. Thus only symetric modes contribute to the effective interactions between charge carriers. However, as mentioned in the introduction, there is convincing experimental and theoretical evidence[12–14] that a more accurate treatment including the apical oxygen ions could give rise to significant corrections. Indeed, recent experimental findings show that both in-plane and apical oxygen ions are occupied with holes in optimally doped compounds.[14] Including the apical oxygens would require another important oxygen phonon mode to be included, namely, that in which the in-plane oxygen ions moving in one direction (in or out with respect to the central copper ion) with the apical oxygen moving in the opposite direction (out or in, respectively). This is a subject for further research.

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