Electron-lattice interaction in cuprates: Effect of electron correlation

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Strong electron-phonon coupling in the cuprates enhanced by electron correlation is suggested by exact diagonalization of a two-band Peierls-Hubbard Hamiltonian in one dimension. To describe the cuprates the band filling was chosen to be about $\frac{3}{4}$, and the effect of changing the copper on-site repulsion and the charge density was examined. The zone-boundary longitudinal optical phonon modes were found to couple very strongly to holes as a result of charge transfer and changes in the local spin correlation induced by deformation. In particular the oxygen mode was found to soften significantly with doping, in agreement with the experimental observation. These results suggest strong synergetic effects of spin-charge-lattice coupling in this system. Implications of these effects to the superconductivity of the cuprates are discussed. [S0163-1829(97)05205-3]

I. INTRODUCTION

The effects of strong electron correlation in transition metal oxide compounds such as the high-temperature superconducting cuprates have been studied extensively by a large number of researchers.[1] While most of the studies have been made from the magnetic point of view,[2] the importance of the lattice effects has been recognized.[3–8] The phenomena are complex and few analytical means are available to solve the problem particularly near the metal-insulator transition. Numerical techniques, on the other hand, can offer reliable and useful reference points, even though they are limited to small systems in low dimensions.[9] We earlier studied a two-band Peierls-Hubbard Hamiltonian by the exact diagonalization method applied to a small one-dimensional system, and found that at half-filling ferroelectricity is enhanced near the crossover condition for many-body states.[10–13] For instance, when the lower Hubbard level of a transition metal ion, such as Ti, is close to the upper Hubbard level of an oxygen ion, ferroelectric lattice deformation induces charge transfer and local spin-resonance between Ti and O, and results in a strongly enhanced electron-lattice interaction favoring ferroelectricity. This is because lattice deformation affects not only the single electron band energy but also the spin exchange interaction through the on-site electron repulsion energy. The total change in the ground state energy can be much greater than for the noninteracting electron system.

A similar enhancement of electron-lattice interaction due to deformation-induced charge transfer is expected to occur in the cuprates which can have profound implications for superconductivity.[10] However, it is more difficult to apply the exact diagonalization technique on the cuprates since the superconductivity occurs at a relatively low level of doping ($\sim$15%) and a simulation of this doping effect suffers from significant size effects. In order to shed some light to this difficult problem in this paper we first vary parameters that affect the excitation gap and then change the electron density to examine the effect of hole doping.

II. METHOD

We describe electrons in a binary p-d system, where d represents the d orbital of a transition metal ion such as Cu^{2+} and p is the p orbital of oxygen, by a one-dimensional two-band Hubbard Hamiltonian;

$$H=\sum_{i,\sigma}\varepsilon_d a_\sigma^+(i)a_\sigma(i)+\sum_{j,\sigma}\varepsilon_p b_\sigma^+(j)b_\sigma(j)+\sum_i U_d n_{d\uparrow}(i)n_{d\downarrow}(i)+\sum_j U_p n_{p\uparrow}(j)n_{p\downarrow}(j)+\sum_{ij} t_{ij}[a_\sigma^+(i)b_\sigma(j)+b_\sigma^+(j)a_\sigma(i)], \tag{1}$$

where

$$n_{d\sigma}(i)=a_\sigma^+(i)a_\sigma(i)$$
$$n_{p\sigma}(j)=b_\sigma^+(j)b_\sigma(j) \tag{2}$$

and i and j label d and p atoms, respectively, σ denotes spin $a_\sigma^+(i)$ is an electron creator on the d atom at site i, $b_\sigma^+(j)$ is an electron creator on the p atom at site j, ε_d, and ε_p are the

single site energy of the d and p state. We assume that the transfer matrix t_{ij} is nonzero only for the nearest-neighbor pairs, and depends upon the atomic displacement as

$$t_{ij}=t_{ij}^{0}\left[1-\alpha(u_i-u_j)\frac{R_i-R_j}{|R_i-R_j|}\right], \tag{3}$$

where u_i is the displacement of the ith ion in the unit of the cell length, R_i denotes the position of the ith ion in space, and α is the electron-lattice coupling constant.

In order to loosely represent the cuprates, we choose the following parameters:

$$\varepsilon_d=0,$$

$$\varepsilon_p=2\,\text{eV},$$

$$t^0 \ \ (\text{for the nearest-neighbor Cu-O pair})=1\,\text{eV},$$

$$\alpha=1,$$

$$U_d=8\,\text{eV} \ \ (\text{in the undoped state}),$$

$$U_p=4 \ \ \text{eV},$$

and initially assume $\frac{3}{4}$ filling, or 3 electrons per unit cell. In the undoped state the hole is mostly on the d site (Cu), except for some leakage to the p site (O) due to hybridization. The lattice effect on this system will be studied at first by varying the value of U_d. As a consequence of doping holes spend more time on oxygen sites, thus the *effective* on-site repulsion energy should be reduced. Thus varying U_d is expected to produce similar, if not the same, effect as doping holes.

It is useful to consider the effect of varying the value of U_d first by neglecting the hopping term, t_{ij}. When the value of U_d is sufficiently large the upper Hubbard state of the d orbital is higher than that of the p orbital, and the hole occupies the d upper Hubbard level. The electron configuration can be denoted d^1p^2. For the cuprates this corresponds to the $Cu^{2+}O^{2-}$ state. If the value of U_d is reduced, at

$$U_d=U_{c0}=U_p+(\varepsilon_p-\varepsilon_d)=6 \ \ \text{eV} \tag{4}$$

the two upper Hubbard levels crossover. When U_d becomes less than U_{c0} the hole moves to the p site. This state is the $Cu^{+}O^{-}$ state, denoted as d^2p^1. If we include the hopping term, the levels develop into bands and the crossover point becomes a continuous range. As U_d is reduced from a large value the system first undergoes a phase transition at U_{uc} (upper critical point) by the delocalization of holes from the d site. The system is most likely metallic when it is in this parameter range. When the value of U_d is further reduced it becomes a Mott insulator again at U_{lc} (lower critical point) if the bandwidth is smaller than U_p. In the real cuprates the value of U_p would not be large enough to drive the system insulating when the value of U_d is small, and the system would remain metallic. However, in this work we assumed otherwise in order to illustrate the effect of the lattice coupling to the holes on oxygen ions. The schematic electronic density of states is shown in Fig. 1 for these three regimes. It is of interest to investigate the effect of lattice distortions (phonons) near such transitions. The Hamiltonian (1) was

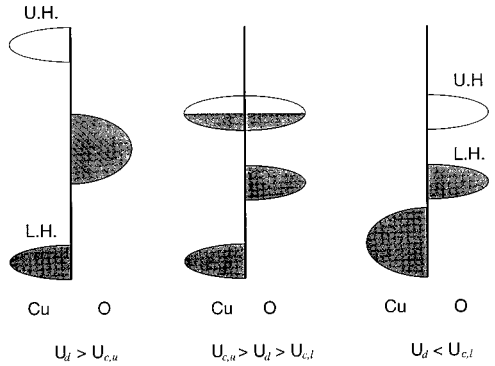

FIG. 1. Schematic one-electron density of states for three parameter regions. (a) $U_d > U_{\text{uc}}$, holes are in the copper upper Hubbard band, (b) $U_{\text{uc}} > U_d > U_{\text{lc}}$, the system is metallic, (c) $U_{\text{lc}} > U_d$, holes are in the upper Hubbard band of oxygen.

numerically diagonalized using the Lanczos method[14,15] for finite-size one-dimensional systems composed of $4d$ and $4p$ sites (4+4 system), $6d$ and $6p$ sites (6+6 system), as well as $8d$ and $8p$ sites (8+8 system). As a boundary condition we used both the simple periodic boundary condition (a ring without a twist) as well as the π-twisted torus condition, in which the phase of the wave function is changed by π after going around the ring once. The total spin was set to be zero. The ground-state energy, the one electron excitation energy, local charges, and spin correlation functions were calculated using a supercomputer, HITAC S-3800.

III. RESULTS

The nature of the phase transitions associated with the level crossover were studied by calculating the charge transfer excitation gap, E_g, and the second-nearest-neighbor spin correlation, $K_2=\langle \mathbf{S}_i\cdot\mathbf{S}_{i+1}\rangle$, for Cu and O for the undeformed structure as a function of U_d. The second-nearest-neighbor spin correlation, K_2, shown in Fig. 2 exhibits discontinuities at two values of U_d, $U_{\text{lc}}=4.7$ eV for O spin correlation and at $U_{\text{uc}}=6.9$ eV, for Cu spin correlation, suggesting phase transitions. The intermediate phase develops around the crossover condition for the case of $t=0$, $U_{c0}=6$ eV, as expected. The excitation gap e_g was evaluated by calculating the one electron excitation energy,

$$E_g=E_0(N+1)+E_0(N-1)-2E_0(N), \tag{5}$$

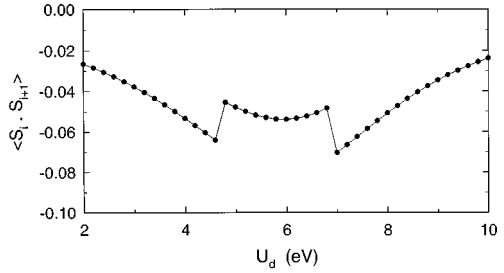

FIG. 2. Dependence of the second-nearest-neighbor spin correlation $K_2=\langle \mathbf{S}_i\cdot\mathbf{S}_{i+l}\rangle$ on U_d calculated for the 4+4 system.

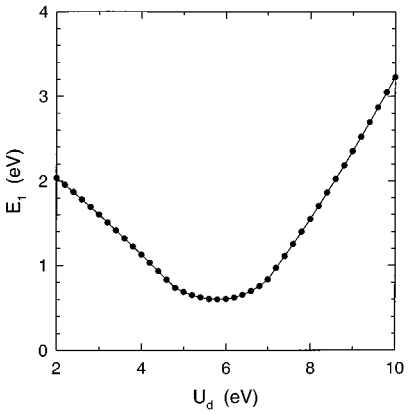

FIG. 3. U_d dependence of the one electron excitation energy calculated for the 4+4 system.

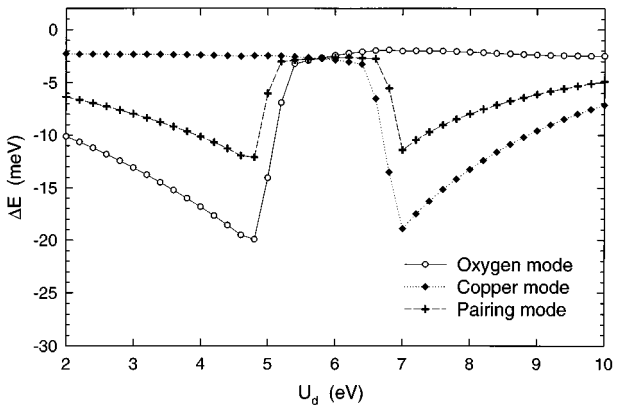

FIG. 5. Energy change per unit cell due to distortion, ΔE, for the 4+4 system at $k=\pi$, for the modes (b)–(d).

where $E_0(N)$ is the ground-state energy with N electrons. The excitation gap does not close as expected for the metallic state, but merely becomes minimum around U_{c0} as shown in Fig. 3. This could be due to the size effect, but it is also possible that this intermediate state is not truly metallic, and is a strange metal or strange insulator, due to a fairly large magnitude of U_p. While this point needs further investigation, this distinction does not appear to be so important to the effect being discussed here.

In order to evaluate the strength of the electron-lattice interaction we then assumed periodic lattice distortions in the frozen phonon approximation. The change in the electronic ground-state energy per unit cell, $\Delta E=[E(u)-E(0)]/L$, where L is the number of unit cells in the system, was calculated for various longitudinal optical phonon modes,

$$u_i=u_d\cos(kR_i+\delta),$$
$$u_j=u_p\cos(kR_j+\delta), \tag{6}$$

where $R_i=i$, $R_j=j+\frac{1}{2}$, δ is the phase of the mode and the unit-cell constant is taken to be unity. As shown in Fig. 4, the zone-center ($k=0$) mode is the ferroelectric mode (a). As the for the zone-boundary ($k=\pi$) modes we consider three types of breathing or antiferroelectric modes depending on the choice of δ; (b) oxygen-displacing mode (Cu breathing mode), (c) Cu-displacing mode (oxygen breathing mode), and (d) the Cu-O pairing mode for which both Cu and O are displaced. While (d) is merely a linear combination of (b) and (c), because of the expected strong nonlinearity[10–13] this mode will be included in addition. The lattice energy was not considered here, since our purpose is to evaluate the strength of the interaction rather than to determine the equilibrium distortion of the electron-lattice system. However, in order to make comparisons of different modes meaningful the rms value of the relative atomic displacements, $\langle(u_i-u_j)^2\rangle^{1/2}$, where i and j are the nearest neighbors, was kept constant at 0.1.

In many cases the electronic energy was found to decrease as the lattice was distorted with the optical phonons. The energy changes due to the distortion, $\Delta E=[E(u)-E(0)]/L$, are shown in Fig. 5 for various modes at $k=\pi$ for the 4+4 system. As in the case of earlier calculations significant decrease in the energy was observed near the critical values of U_d. The oxygen displacing mode results in a minimum in ΔE at 4.9 eV, while the copper displacing mode produces a mirror image with a minimum at 7 eV. The values of U_d corresponding to the two minima are close to the transition points observed for the spin correlation shown in Fig. 2, suggesting that the maxima in the electron-lattice coupling are associated with the phase transitions. Note that below $U_{\rm lc}$ hoes are mainly on oxygen, while above

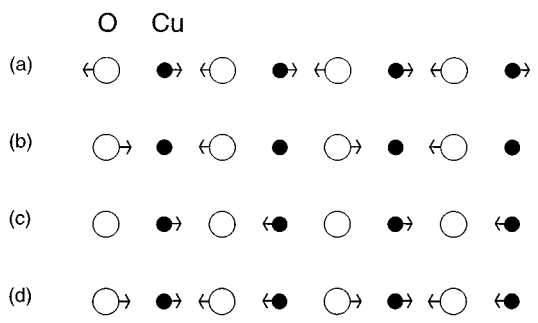

FIG. 4. LO phonon modes, (a) ferroelectric mode, $k=0$; (b) zone-boundary oxygen displacing (copper breathing) mode, $k=\pi$; (c) zone-boundary copper displacing (oxygen breathing) mode, $k=\pi$; (d) zone-boundary pairing mode, $k=\pi$.

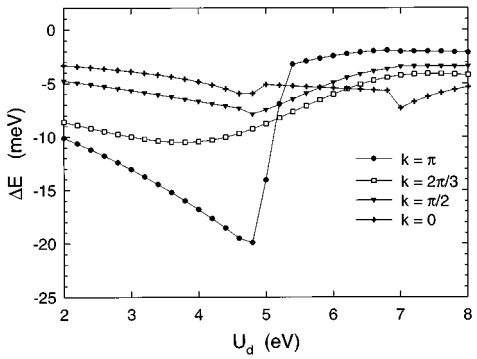

FIG. 6. Wave-vector dependence of ΔE, evaluated at various values of k as a function of U_d for the oxygen displacing modes. The result for $k=2\pi/3$ is for the 6+6 system. Others are for the 4+4 system.

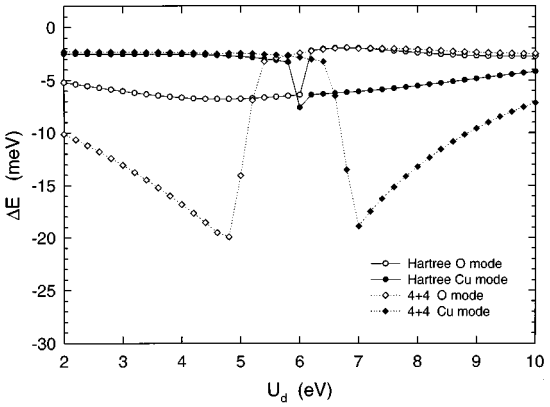

FIG. 7. Energy change per unit cell due to distortion (b) and (c), ΔE, calculated in the Hartree approximation and compared with the results in Fig. 5.

$U_{\rm uc}$ they are mainly on Cu. In agreement with Ref. 8 the mode which pairs up holes are softened when t is modulated by deformation. The Cu-O pairing mode leads to two minima at these critical values. The energy change for the mode (d) is slightly larger than the average of those for (b) and (c), underscoring the nonlinearity of the phenomena. A similar calculation was carried out for the case of $\varepsilon_p=\varepsilon_d$. The result was almost identical except that the crossover point was shifted by 2 eV. Since the system is truly metallic at least when $U_d=U_p$, this result appears to suggest that the question whether the intermediate phase between $U_{\not c}$ and $U_{\rm uc}$ is truly metallic or not is immaterial to this effect.

The amount of energy change is strongly dependent upon k as shown in Fig. 6. Here ΔE is shown as a function of U_d for the oxygen displacing modes at several values of k. The result for $k=2\pi/3$ was obtained for the 6+6 system, while others were calculated for the 4+4 system. Clearly only the zone-boundary $k=\pi$ mode most strongly reduces the electron energy. This should be contrasted to the half-filling case we studied earlier where zone-center $k=0$ ferroelectric mode showed a maximal change. This suggests that the maximum electron-lattice interaction occurs at or near $k=2k_F^0$ where k_F^0 is the Fermi wave vector of the corresponding noninteracting electrons. Indeed for $\frac{2}{3}$ filling the maximum interaction was observed to occur at $k=2\pi/3$. It may thus appear that the role of U_d in the observed enhancement of the electron-lattice coupling is merely to bring about the metallic behavior; when the common upper Hubbard band for d and p is formed and the metallic behavior is realized, the usual Fermi surface nesting mechanism goes at work. However, if that is the case the coupling should be maximum at the middle of the intermediate region, around $U_{c0}=6$eV. Instead the maximum interaction occurs sharply at the two transition points, where the charge transfer gap is small. In the middle of the intermediate (metallic) region the coupling is weak. Furthermore, as we mentioned above whether the intermediate region is truly metallic or not is unclear. Therefore this strong interaction must be closely related to the phase transition itself, rather than to the Fermi surface nesting. In this sense we consider this phenomenon represents *unconventional* electron-lattice coupling.

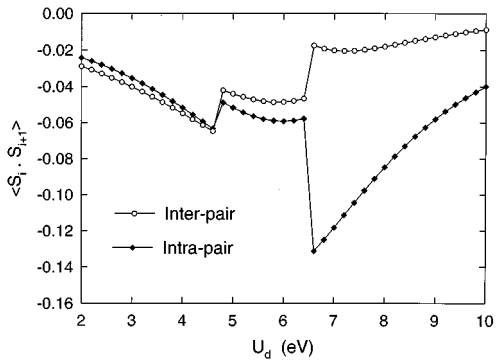

FIG. 8. U_d dependence of the nearest-neighbor Cu-O spin correlation, $K_1=\langle \mathbf{S}_i\cdot\mathbf{S}_j\rangle$ with the copper displacing mode, Fig. 6(c).

In order to assess the effect of electron correlation on the magnitude of this effect we also calculated the coupling energy in the Hartree approximation,

$$Un_\uparrow n_\downarrow \Rightarrow U(\langle n_\uparrow\rangle n_\downarrow+\langle n_\downarrow\rangle n_\uparrow-\langle n_\uparrow\rangle\langle n_\downarrow\rangle) \tag{7}$$

and assuming that $\langle n_{i,\sigma}\rangle=\langle n_{i+2,\sigma}\rangle$ which promotes antiferromagnetic order. The changes in the energy thus calculated are shown in Fig. 7. Compared to the results in Fig. 5 the energy change is much smaller, by more than a factor of 3, and is only weakly dependent on U_d. This result clearly indicates the importance of electron correlation which is neglected in the Hartree approximation. The strength of electron correlation is seen in the nearest-neighbor Cu-O spin correlation $K_1=\langle \mathbf{S}_i\cdot\mathbf{S}_j\rangle$ shown in Fig. 8. The intrapair K_1 for the Cu-O pair for which t is increased by deformation becomes sizable in magnitude near the phase boundary. Its dependence on U_d is similar to that of the energy and $J(J\sim t^2/U$ for large $U)$, suggesting that this spin correlation is the principal reason for the large energy gain. The interpair spin correlation is reduced by the deformation. As a result of this change the superexchange interaction is modified, resulting in large changes in the second-neighbor (Cu-Cu or O-O) spin correlation as shown in Fig. 9.

The deformation leads also to the formation of a charge density wave (CDW). As shown in Fig. 10, when U_d is large a pair of Cu atoms which move closer to each other by deformation increases the hole density on the oxygen atom in between, and create a CDW on oxygen sublattice. When

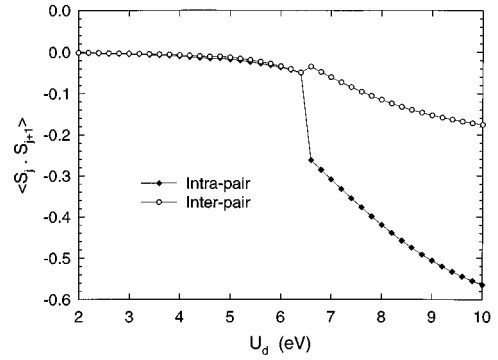

FIG. 9. U_d dependence of the second-nearest-neighbor copper spin correlation, $K_2=\langle \mathbf{S}_i\cdot\mathbf{S}_{i+1}\rangle$, with the copper displacing mode, Fig. 6(c).

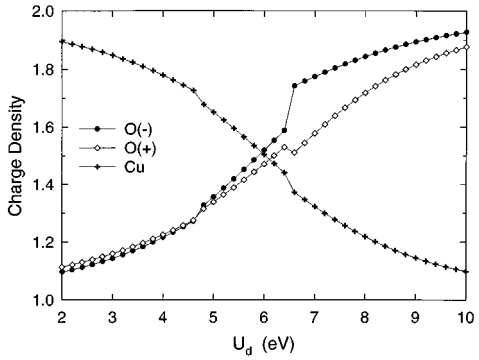

FIG. 10. U_d dependence of the charge densities on oxygen squeezed by a pair of copper atoms O(+), on oxygen between the pair O(−), and on copper, when the copper displacing mode [Fig. 6(c)] is activated.

U_d is smaller that U_{lc}, an almost exact mirror image is formed. The CDW is produced on Cu by the oxygen displacing mode, and the O-O spin correlations and the Cu-O spin correlations are modified as expected.

In order to evaluate the effect of the system size calculations were performed also for the π-twisted boundary condition and for the 6+6 as well as 8+8 systems. It was found that the results for three systems, the 4+4, 6+6 with the π twist, and 8+8 systems, which we call Group A, are very similar as shown in Fig. 11, except for slight differences in the critical values of U_d. On the other hand the 4+4 system with the π twist and the 6+6 system with the regular boundary condition, Group B, showed no sharp features with shallower minima, as shown in Fig. 12. The minima for the 6+6 system, however, are significantly deeper than those of the 4+4 system with the π twist. It is likely that the abruptness of the changes at the phase boundaries for the Group A is a size effect in one-dimensional systems as discussed in Ref. 16. In fact our preliminary results on two-dimensional systems show similar, but more symmetric, V-shaped minima. The depth of the minima, however, remains practically unchanged with the size for the Group A. The depth of the minima for the Group B appears to converge toward the

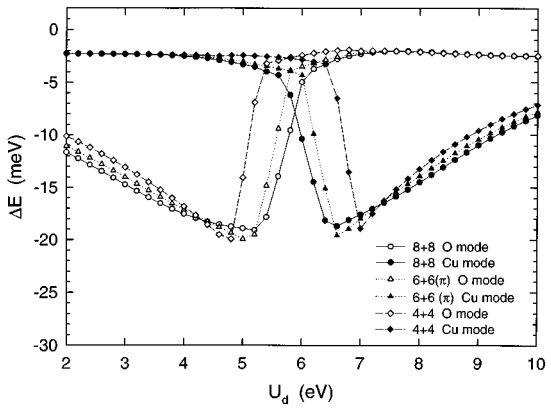

FIG. 11. Energy change per unit cell due to distortion (b) and (c), ΔE, for the 4+4 system, the 6+6 system with the π-twisted boundary condition, and the 8+8 system with the normal boundary condition.

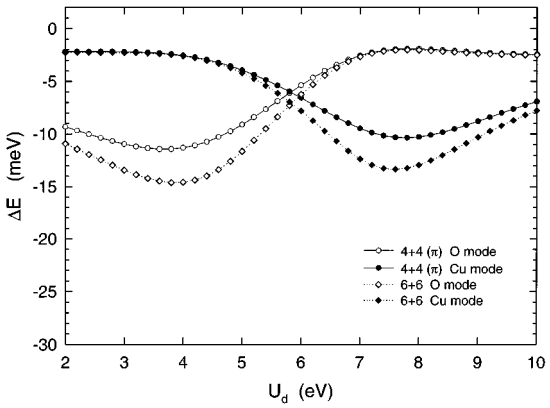

FIG. 12. Energy change per unit cell due to distortion (b) and (c), ΔE, for the 4+4 system with the π-twisted boundary condition, and the 6+6 system with the normal periodic boundary condition.

values for Group A as the sample size is increased. Therefore it is most likely that the depth of the minima is correctly represented by the results of the Group A while the abruptness of the transition is exaggerated, although further studies are necessary on this point.

Similar calculations were performed with hole doping. The results for the 6+6 system with two and four extra holes added are shown in Fig. 13 for the case of the oxygen displacing mode (b), and in Fig. 14 for the Cu displacing mode (c). When U_d is large as in the cuprates the oxygen displacing mode is strongly softened by doping, while the Cu displacing mode is hardened. For a typical value of U_d in cuprates, 10 eV, this results in the oxygen LO phonon softening of the order of 10 meV due to hole doping of 33%. Due to the size effect, however, the present result may not accurately reflect the real magnitude. It is interesting to note that the amount of softening is about the same for the case of two hole doping ($\frac{1}{3}$ doping) and the case of four hole doping ($\frac{2}{3}$ doping) for the oxygen mode (Fig. 13), and is even reversed in the case of the Cu mode, indicating the strong softening occurs only for relatively low doping levels. This

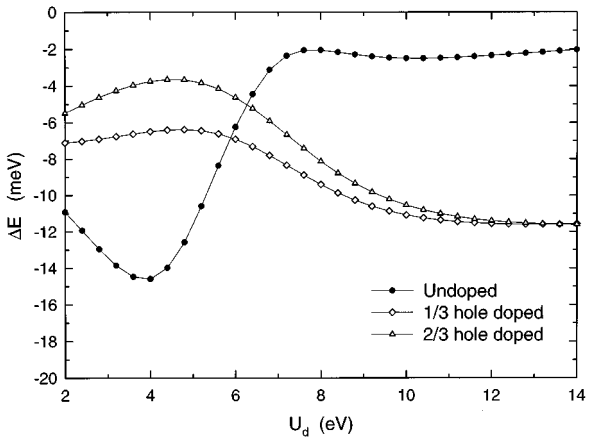

FIG. 13. Energy change per unit cell due to oxygen mode distortion (b), ΔE, for the 6+6 system with the periodic boundary condition, for the undoped system with 18 electrons ($\frac{3}{4}$ filling), the doped system with two extra holes (16 electrons, $\frac{1}{3}$ doping), and with four extra holes (14 electrons, 2/3 doping).

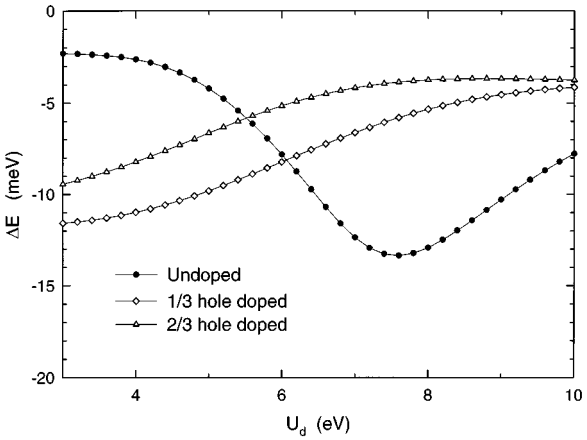

FIG. 14. Energy change per unit cell due to copper mode distortion (c), ΔE, for the 6+6 system with the periodic boundary condition, for the undoped system with 18 electrons ($\frac{3}{4}$ filling), the doped system with two extra holes (16 electrons, $\frac{1}{3}$ doping), and with four extra holes (14 electrons, $\frac{2}{3}$ doping).

tendency of initial rapid softening with doping and slow hardening afterward is similar to the results of the crossover in the undoped system shown in Figs. 5 and 11. Therefore the system with the value of U_d just below $U_{\rm lc}$ could loosely represent the doped cuprates in their behavior.

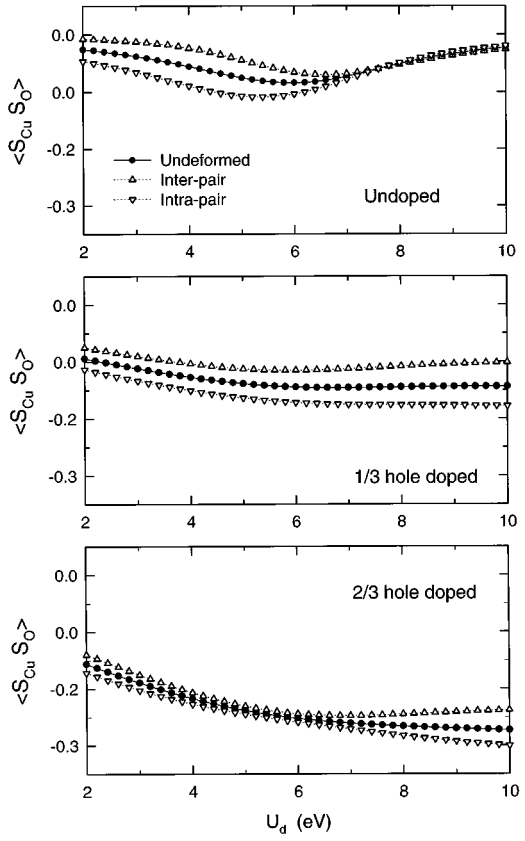

FIG. 15. Variation of the Cu-O spin correlation as a function of U_d, undeformed (closed circle), deformed with the oxygen displacing mode (b), for the intrapair spins between which t is increased (open downward triangle) and interpair spins between which t is decreased (open upward triangle).

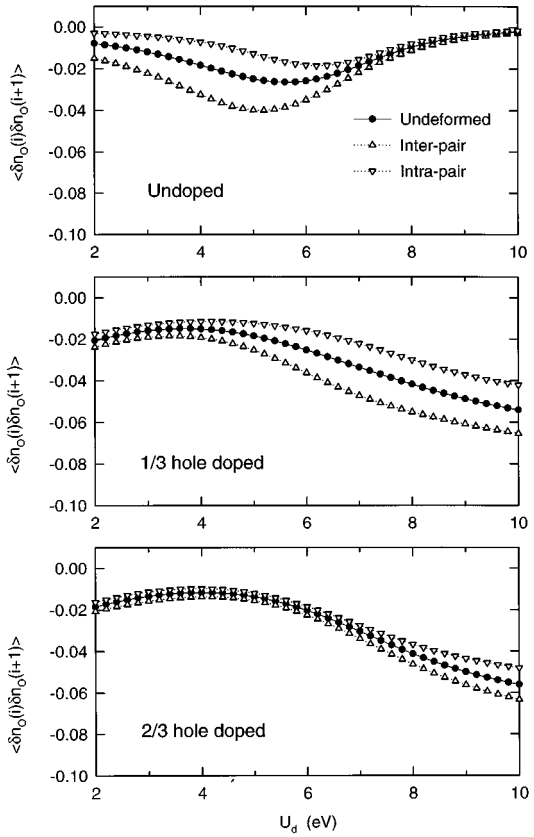

FIG. 16. Variation of the O-O charge density correlation as a function of U_d, undeformed (closed circle), deformed with the oxygen displacing mode (b), for the intrapair and interpair oxygen ion correlations (open triangles).

The strong softening is due to the increased Cu-O spin correlation as shown in Fig. 15. Again the effect of deformation is much smaller for the system with $\frac{2}{3}$ doping. Charge correlation between the nearest oxygen neighbors is shown in Fig. 16. The result indicates that holes repel each other as expected. This result does not necessarily imply that there is no tendency to form static bipolarons, because the lattice is frozen and is not involved in the energy minimization in this work. The interaction between holes mediated by the lattice will be studied in the future works. Since the zone-boundary LO phonon increases the Cu-O exchange constant $J_{\rm pd}$ for a half of the Cu-O pairs and decreases it for the other half, the effective Cu-Cu exchange is almost unchanged as indicated in terms of the Cu-Cu spin correlation shown in Fig. 17.

IV. DISCUSSION

A. Comparison with experimental results

The results above predict softening of the zone-boundary LO phonon due to strong electron-phonon interaction. Indeed the softening of such a mode has been observed for $YBa_2Cu_3O_{7-\delta}$,[17] $La_{2-x}Sr_xCuO_4$,[17] and $(Ba_{1-x}K_x)BiO_3$ (Ref. 18) by neutron inelastic scattering as an insulating parent compound is doped with holes. The softening is most pronounced for the oxygen displacing mode at the $(\pi,0)$ point (zone boundary in the direction of the Cu-O bond) in all three cases. In $La_{2-x}Sr_xCuO_4$ the energy of the $(\pi,0)$

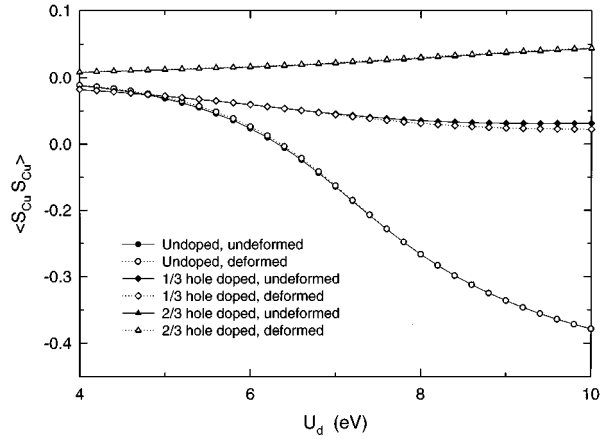

FIG. 17. Variation of the Cu-Cu spin correlation as a function of U_d undeformed (closed symbol) and with the oxygen displacing mode (b) (open symbol) for undoped, $\frac{1}{3}$ hole doped, and $\frac{2}{3}$ hole doped systems.

mode is reduced from 19.2 THz (79 meV) for $x=0$ to 17.4 THz (72 meV) for $x=0.1$,[17] and 70 meV for $x=0.15$.[19,20] In $YBa_2Cu_3O_{7-\delta}$ the mode softens from 18.5 THz (77 meV) for $\delta=1$, to as low as about 14 THz (58 meV) for $\delta=0$.[17] In highly oxygenated $YBa_2Cu_3O_{7-\delta}$ the LO mode is mixed with other modes with similar frequency. In $Bi_2Sr_2(Ca_{1-x}Y_x)Cu_2O_8$ as x is changed from 1 ($T_c<4$ K) to 0 ($T_c\sim 80$ K) the LO modes in the range of 50 to 80 meV softens appreciably, although in this case the wave vector dependence has not been resolved.[21]

The phonon softening in $YBa_2Cu_3O_7$ has been explained in terms of the Fermi surface nesting.[22] However, in $La_{2-x}Sr_xCuO_4$ the Fermi surface is rotated by 45° compared to $YBa_2Cu_3O_7$ and indeed the calculated phonon susceptibility is sharply peaked around the (π,π) point, with almost no intensity at the $(\pi,0)$ point.[23] Therefore it is impossible to explain *both* by the Fermi surface nesting. Phenomenologically the softening of the oxygen LO mode at $(\pi,0)$ implies the presence of an *attractive* interaction, or a negative force constant between oxygen atoms. Since oxygen ions repel each other electrostatically, it is not easy to explain this attraction by classical mechanisms. The mechanism discussed here provides a natural explanation of this attractive potential in terms of the spin exchange interaction. Indeed the observed magnitudes of the phonon softening are similar to the results shown above. Our results also predict *hardening* of the Cu displacing mode at $(\pi,0)$. Unfortunately the Cu LO mode is mixed up with other modes with similar energies, and is not easy to identify. Experimental verification of this point is of great interest.

Since the present calculation is done for a one-dimensional system, it is not possible to differentiate the effect of the $(\pi,0)$ half-breathing phonon from that of the (π,π) breathing phonon. However, the $(\pi,0)$ phonons propagate along the Cu-O bond and thus retains a one-dimensional character. The (π,π) phonons produce charge changes in the nearest-neighbor oxygen sites, so that if we introduce Coulomb repulsion and hopping between them, the (π,π) mode will be less favored, as indicated by our recent calculations on two-dimensional systems which will be reported separately.

It is interesting to note that a $(\pi,0)$ charge ordering is likely to be present in $La_{2-x}Ba_xCuO_4$ with $x=\frac{1}{8}$.[24] Superconductivity is suppressed in this compound near the $\frac{1}{8}$ composition[25] probably due to charge ordering. According to the recent magnetic neutron-scattering measurement on $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$,[24] at low temperatures charges are localized on every fourth row of Cu-O chain in the (1 0 0) direction in the plane, with the charge density of $\frac{1}{2}$ within the row, forming stripes. While the nature of charge ordering within the row is not yet known, the charge density of $\frac{1}{2}$ is suggestive of the $(\pi,0)$-type charge ordering. It is possible to imagine that the holes are always dressed with the $(\pi,0)$ half-breathing mode as suggested by the present calculation, and when the periodicity corresponding to the average charge density is commensurate with the $(\pi,0)$ mode, i.e., when the average charge density is rational, static charge density wave is formed and pinned by the lattice.

Another signature of this coupling mechanism is the strong nonlinearity of electron-lattice coupling.[10–13] Near the crossover point the lattice deformation induces large charge transfer, drastically changing the electronic ground state, and this change is the origin of such a strong interaction. Consequently the response is quite nonlinear. This nonlinearity is expected to cause strong anharmonicity in the lattice. Indeed while the lattice anharmonicity evaluated by the LDA calculations is not strong,[26,27] various lattice anomalies indicating very substantial lattice anharmonicity have been observed.[28] As for the LO modes the neutron-inelastic-scattering results on $La_{2-x}Sr_xCuO_4$ (Ref. 17) indicate that the phonon lifetime is anomalously short around the $(\pi,0)$ point, while even within the same branch the energy width of the (0,0) point phonon is resolution limited. Phenomenologically this q dependence of the energy width implies that the Cu-O potential is harmonic, but the O-O attractive interaction is anharmonic, which is exactly what the present mechanism predicts. A recent neutron-inelastic-scattering measurement[19] showed a strong asymmetry in the peakshape as a function of energy transfer for the $(\pi,0)$ LO mode, which further attest to the unusual nature of this mode.

There is no apparent mark of strong electron-lattice coupling on the electronic structure itself. However, the extended saddle point which has been observed in all the superconducting cuprates studied by photoemission so far[29,30] except for electron-doped $Nd_{2-x}Ce_xCuO_4$ may be indicative of this coupling. Bulut *et al.*[31] explained this phenomenon in terms of the many-body spin-polarons, but an alternative explanation is possible with the phonon dressing. It should be noted that the extended saddle point extends along Δ almost exactly halfway from the original saddle point (X or M). The nearly complete absence of dispersion indicates that the states are already localized, or can readily be localized, by forming a wave packet. Such wave packets produced by summing the Bloch states from $\pi/2$ to $3\pi/2$ are the Wannier functions with the width twice the usual Wannier function and located at alternating sites. They can couple strongly to the $(\pi,0)$ LO modes discussed here. It is therefore possible that this coupling is the origin of the flatness of this extended saddle point, by inducing these electrons to localize, or at least to be heavily dressed by phonons. They may possibly be dressed by paramagnons at the same time.

The nonlinearity of the coupling and strong electron correlation are likely to result in formation of small local domains or regions where wave packets of electrons and phonons reside in a palaronic state, just as in the correlation bag proposed by Goodenough.[5] In fact various local structural probes indicate the presence of such local structural variations.[28] Holes on oxygen atoms are more likely found in this region. However, at this moment we do not know how many holes reside in the region, and cannot conjecture if this domain will be a polaron, a bipolaron, or merely a region of segregated holes.[32] It is possible that these regions take a form of stripes as conjectured by Zaanen and Gunnarson.[33] In the charged region the Cu hole density will be slightly reduced, thus electrons are transferred from oxygen to copper. This would result in the weakening of the Cu-O bond and local extension of the Cu-O bond length, but since the charged region is constrained by the matrix which is in the underdoped state, the Cu-O-Cu bond will be buckled with the in-plane oxygen displaced along the c axis[5] and the apical oxygen will move closer to copper. This explains the anomalous local atomic displacements of oxygen atoms along the c axis that are frequently observed by local structural probes[28,34] and the strong coupling between the c-axis phonons and the in-plane charge dynamics.[35] Outside the charged region the hole density is low and local Cu spin coupling will be strong. The charge carriers in the strongly coupled regions would behave quite differently from the nearly free carriers on the fast dispersing portion of the Fermi surface, resulting in two kinds of charge carriers. In fact the presence of two kinds of carriers has been suggested by the IR absorption[36] and NMR[37,38] measurements, as well as theories.[39,40]

It is interesting to note that special care must be exercised to describe the crossover effects and the zone-boundary LO phonon softening discussed here by the prevailing one-band model.[41,42] For instance in the t-J model the phonon modulation of t and J would not describe the present effects. Indeed as shown in Fig. 17 the zone-boundary LO phonon hardly affects the Cu-Cu spin correlation. The LO phonon would couple to the local level of Cu, and this coupling should depend upon the total charge density.

B. Relevance to superconductivity

The strong electron-lattice coupling predicted by the present calculation invites a speculation that this interaction may form a basis for lattice mediated superconductivity in the cuprates. The idea is even more attractive in the light of the recent results of inelastic-neutron-scattering measurements which place severe restrictions on the strength of magnetic excitation[43] or the spatial extension of magnetic correlation[44] both of which are necessary for the spin-fluctuation mechanism[45–47] to explain the high-temperature superconductivity (HTSC) phenomena successfully. For instance the $(\pi,0)$ LO phonon modes are expected to couple strongly to the Wannier functions made of the states in the extended saddle point as we discussed above, and could create hole pairs. Indeed the superconducting gap is maximum in the direction of the extended saddle point. While this possibility remains purely a speculation at this moment it would be useful to discuss what properties the cuprate superconductor is likely to exhibit if this mechanism is indeed at work.

It is well known that the high-temperature superconductivity arises near the phase boundary between the Mott insulator and a metal.[48] Indeed the increases in the low-temperature dc conductivity as a function of charge concentration extrapolated from the high-temperature conductivity[49] and optical conductivity[50] and that in the electronic specific heat[51] coincide with the maximum in T_c. Varma argued that the interband charge transfer excitons which become strong as the pseudogap narrows could provide the pairing force.[52] The present mechanism naturally explains this close connection between the superconductivity and the Mott-insulator-to-metal transition. In other mechanisms, however, the correspondence between the metal-insulator transition and the maximum in T_c is coincidental. For instance in the spin-fluctuation mechanism[45–47] T_c increases as the magnetic fluctuation softens, and then T_c decreases as the magnetic correlation is further weakened. In this picture the coincidence of the disappearance of antiferromagnetism and metal-insulator transition is largely accidental, even though the metallic behavior certainly disrupts antiferromagnetic order.

Another possible characteristic of the superconductor due to the electron-phonon interaction enhanced by electron correlation is that the isotope effect must be small, since most of the screening is achieved by electronic polarization. As shown in Refs. 10–13 and confirmed by Resta and Sorella[16] the effective charge associated with the deformation is much larger than the real charge. This is because the lattice polarization P induces large electronic polarization p through the deformation-induced charge transfer. Consequently the isotope effect should be minimum when T_c is maximum, since the electronic component of screening is maximum there. These agree with the experimental observations which are difficult to explain otherwise.[53] Furthermore it is possible to imagine that the d-wave superconductivity would result from this mechanism. Even though the issue of the symmetry of the order parameter is still controversial,[54–59] the d symmetry has always been cited as an argument against the phonon mediated superconductivity. However, d-wave superconductivity may result from electron-phonon coupling with the background of antiferromagnetism.[60] While the BCS mechanism prefers the s-wave, in the present case coupling is achieved by LO phonons with a special symmetry, preferring x or y direction. The attractive phonon-mediated interaction near the $(\pi,0)$ point and the repulsive interaction due to magnetic fluctuation near the (π,π) point could lead to the d-wave superconductivity.

V. CONCLUSION

The effect of lattice distortion on a strongly correlated electron system such as the cuprates was studied by exact diagonalization of the two-band Peierls-Hubbard Hamiltonian. The band filling was chosen to be near $\frac{3}{4}$, corresponding to the electron concentration in the high-temperature superconducting cuprates. The electron-lattice coupling was studied first by varying the strength of the copper on-site repulsion and then actually introducing doped holes in the system. The result shows that at small doping or near the transition from the Mott insulator to a metal the zone-boundary longitudinal optical phonon modes couple very

strongly to electrons and significantly reduce the total energy of electrons. This effect is produced as a result of deformation-induced charge transfer and deformation-induced spin coupling. We propose that this effect explains the pronounced softening of the $(\pi,0)$ LO phonon by doping which has been observed for $YBa_2Cu_3O_{7-\delta}$, $La_{2-x}Sr_xCuO_4$, as well as $(Ba_{1-x}K)_xBiO_3$. If this coupling were to produce superconductivity it would provide a natural explanation for some prominent characteristics of the HTSC phenomena, such as the fact that T_c is maximum while the isotope effect is minimum at the doping level corresponding to the Mott-insulator-to-metal transition. This mechanism could be seen as a *magnetic* mechanism in a wider sense since the local dynamic spin correlation is providing much of the driving force. However, the lattice (phonon) is crucial as the trigger mechanism in the present scheme. Also it should be noted that the static or quasistatic spin correlation would suppress this mechanism, in contrast to the widely discussed spin-fluctuation mechanism. The main thrust of the present mechanism is the concerted effect of lattice, spin, and charge, all conspiring to pair up holes.

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