Asymmetric electron-phonon interactions in the three-band Peierls-Hubbard model

Z. B. Huang, W. Hanke, and E. Arrigoni
Institut für Theoretische Physik, Universität Würzburg, am Hubland, 97074 Würzburg, Germany

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Using the Quantum Monte Carlo (QMC) technique within frozen-phonon, we studied the effects of the half-breathing \(O(\pi, 0)\) phonon mode on the ground-state properties of the three-band Peierls-Hubbard model. Our simulations are performed for both ionic and covalent electron-phonon couplings. The effects of lattice displacements on the ground-state energies and charge fluctuations are similar in magnitude for both hole- and electron-doped cases. However, the effects of lattice displacements on the magnetic properties are rather different. In the hole-doped case, the normalized next-nearest-neighbor Cu-Cu spin correlations are dramatically modified by both ionic and covalent electron-phonon couplings. On the other hand, in the electron-doped case, much smaller effects are observed. The distinct spin-phonon couplings, in conjunction with the spin-bag picture of the quasiparticle, could explain a strong mass renormalization effect in the p-type cuprates and a weaker effect in the n-type cuprates.

Recently, a large number of experimental observations in the superconducting cuprates suggest that the electron-phonon (el-ph) coupling in these systems is strong and -in conjunction with strong electronic correlations, may play a role for the superconductivity mechanism. In particular, angle resolved photoemission spectroscopy (ARPES) data indicate a mass renormalization (“kink”) in the electronic dispersion of the p-type high-\(T_c\) cuprates near a characteristic energy scale (≈ 70 meV)\(^{1,2}\), which is possibly caused by coupling of quasiparticle to some phonon modes. Moreover, ARPES data\(^{3,4}\) also show that this mass renormalization is weak in the n-type Nd\(_{1.85}\)Ce\(_{0.15}\)CuO\(_4\), indicating that the el-ph coupling in the p-type materials is much stronger than in the n-type ones. On the other hand, both inelastic neutron scattering\(^{5,6}\) and inelastic x-ray scattering\(^{7}\) found that the anomalous softening of the Cu-O bond-stretching oxygen mode is present in both p- and n-type materials, suggesting that although there exist some differences, the el-ph coupling is strong in both kinds of materials.

In order to understand this asymmetry of el-ph coupling in p- and n-type cuprates, we have studied the three-band Peierls-Hubbard model in the physically interesting parameter regime by employing the constrained-path Monte Carlo (CPMC) method.\(^{10,11}\) In the CPMC method, the ground-state wave function \(|\Psi_0\rangle\) is projected from a known trial wave function \(|\Psi_T\rangle\) and then the physical quantities are calculated in this ground state. We find that in both hole- and electron-doped cases, the effects of lattice displacements on the ground-state energies and charge fluctuations are similar. However, an essential difference between hole- and electron-doped systems is that the response of spin degree of freedom to phonons is strong in the former case, but rather weak for the latter.

In the hole representation the three-band Peierls-Hubbard model\(^8,9\) has the Hamiltonian,

\[
H = \sum_{\langle i,j \rangle} t^{ij}_{pd}(\{u_j\})(d^\dagger_{i\sigma}p_j + h.c.) + \sum_{\langle j,k \rangle} t^{jk}_{pp}(p_j^\dagger p_k + h.c.) + \epsilon_p \sum_{j\sigma} n_{j\sigma}^p + \sum_{i,i'} \epsilon_{ij}(\{u_j\}) n_{i\sigma}^d + U_d \sum_i n_{i\uparrow}^d n_{i\downarrow}^d + U_p \sum_j n_{j\uparrow}^p n_{j\downarrow}^p + V_{pd} \sum_{\langle i,j \rangle} n_{i\uparrow}^d n_{j\downarrow}^p.
\]

Here the operator \(d^\dagger_{i\sigma}\) creates a hole at a Cu \(3d_{x^2-y^2}\) orbital and \(p^\dagger_{j\sigma}\) creates a hole in an O \(2p_{\sigma}\) orbital. \(U_d\) and \(U_p\) are the Coulomb energies at the Cu and O sites, respectively. \(V_{pd}\) denotes the nearest-neighbor (nn) Coulomb repulsion. For the el-ph coupling, we consider that the nn Cu-O hybridization is modified by the O-ion displacement \(u_j\) as \(t^{ij}_{pd} = \phi_{ij}(t_{pd} \pm \epsilon u_j)\), where the \((+(-)\text{ applies if the bond shrinks (stretches) with positive} \ u_j\). The phase factor \(\phi_{ij}\) takes a minus sign for \(j = i + \hat{x}/2\) and \(j = i - \hat{y}/2\). The Cu-site energy is assumed to be modulated by the nn O-ion displacements \(u_j\) as \(\epsilon_{ij}(\{u_j\}) = \epsilon_d + \beta \sum \pm u_j\). The other electronic matrix elements are the OO hybridization \(t^{jk}_{pp} = \pm t_{pp}\) with the minus sign occurring for \(k = j + \hat{x}/2\) and \(k = j + \hat{x}/2\) and the O-site energy \(\epsilon_p\). The charge-transfer energy is defined as \(\epsilon = \epsilon_p - \epsilon_d\). For simplicity, we introduce dimensionless parameters: \(u = \alpha |u_j|/t_{pd}\), or \(u = \beta |u_j|/t_{pd}\). In units of \(t_{pd}\), we use the set of parameters: \(U_d = 6\), \(\epsilon = 3\), \(U_p = V_{pd} = 0\), and \(t_{pp} = 0.5\).

All the results reported here were done on a lattice with \(6 \times 6\) unit cells and periodic boundary conditions. In the following we will focus on the half-breathing \(O(\pi, 0)\) phonon mode and examine the dependence of physical quantities on the lattice displacement \(u\) in different cases. In order to diminish systematic and statistical errors in the CPMC simulations, we studied two closed-shell cases: the hole doping \(\delta_h = \frac{\epsilon}{3}\) for the hole-doped case and the electron doping \(\delta_e = \frac{\epsilon}{3}\) for the electron-doped case. Notice that we were forced to choose two different dopings for the hole- and electron-doped cases, due to the requirement of a closed-shell condition. However, we...
have checked that the results at a smaller electron doping \( \delta_e = \frac{10}{6} \) on a lattice with 8 \( \times \) 8 unit cells remain qualitatively similar to the ones at \( \delta_e = \frac{3}{4} \), demonstrating that our findings on the 6 \( \times \) 6 lattice reflect intrinsic properties of the three-band Peierls-Hubbard model.

Fig. 1 shows the displacement pattern of the half-breathing \( O(\pi,0) \) phonon mode. The closed circles label the Cu sites, and the open circles the O sites. The arrows denote the lattice displacements from the O sites.

![FIG. 1: Displacement pattern of the half-breathing O(\( \pi,0 \)) mode on the Cu-O plane. The closed circles label the Cu sites, and the open circles the O sites. The arrows denote the lattice displacements from the O sites.](image)

In Figs. 2a, 2b, and 2c, we show the changes of different energies as a function of the lattice displacement \( u \) in the hole-doped case. The corresponding results in the electron-doped case are shown in Figs. 2d, 2e, and 2f. In both hole- and electron-doped cases, the electronic energy is reduced by lattice displacements. As one can see, in the hole-doped case, the covalent and ionic el-ph couplings have an equal effect in the reduction of electronic energy, while in the electron-doped case, the ionic el-ph coupling has a stronger effect than the covalent el-ph coupling.

From Figs. 2b, 2d, and 2f, for the covalent el-ph coupling, one can see that the reduction of electronic energy comes from the gain in kinetic energy, while for the ionic el-ph coupling it originates from the gain in Cu-site potential energy as shown in Figs. 2e and 2f. Notice that although the noninteracting model (\( U_d = 0 \)) also decreases its electronic energy by lattice displacements through the same mechanism, i.e., the gain in kinetic energy or Cu-site potential energy, electronic correlations play an important role in the strongly correlated system. As a matter of facts, for a given lattice displacement \( u \) the Coulomb interaction \( U_d \) transfers some charges from copper sites to oxygen sites, making the kinetic energy \( E_K \) in the interacting system even lower than that in the noninteracting system. This demonstrates that electronic correlations

\[
\begin{align*}
E &= \langle H \rangle, \quad (2) \\
E_K &= \langle \sum_{(i,j)\sigma} t^{ij}_{pd} (\{ u_j \} + d_{j\sigma} p_{j\sigma} + h.c.) + \sum_{(j,k)\sigma} t^{jk}_{pp} (d_{j\sigma} p_{k\sigma} + h.c.) \rangle, \\
E_P &= \langle \sum_{i\sigma} \beta (\sum_j \pm u_j) n_{i\sigma}^d \rangle, \quad (4) \\
E_O &= \langle \epsilon_p \sum_{j\sigma} n_{j\sigma}^p \rangle, \quad (5) \\
E_C &= \langle U_d \sum_{i} n_{i\uparrow}^d n_{i\downarrow}^d \rangle. \quad (6)
\end{align*}
\]

Here, \( \langle \cdots \rangle \) means the ground-state expectation value.
contribute essentially to the gain in kinetic energy. In addition, we observe that for the ionic el-ph coupling and in the noninteracting model, the Cu-site potential energy decreases more in the hole-doped case than in the electron-doped case. On the other hand, Figs. 2a and 2d show that in the strongly correlated system this behavior is reversed, which is consistent with the physical situation where the doped electrons mainly go to the copper sites, as a result, the charge fluctuations at the copper sites are much stronger than those in the hole-doped case.

In general, the strong el-ph coupling in both p- and n-type cuprates can be attributed to the strong interactions of lattice displacements with local metallic charge fluctuations. McQueeney et al. have studied the doping dependence of phonon densities of states in La_{2−x}Sr_xCuO_4 and found that the development of the 70 meV phonon mode near the doping-induced metal-insulator transition is due to the low-energy one-dimensional charge dynamics. Moreover, Ishihara et al. have studied the one-dimensional two-band Peierls-Hubbard model and indicated that the charge-transfer fluctuations are strongly enhanced near the transition from a Mott insulator to an ionic insulator (or a metal). As shown in Fig. 3 due to lattice displacements, the charge densities at both copper and oxygen sites are modified in a similar way for the hole- and electron-doped systems. However, in our cases the charge-transfer fluctuations have little contribution to the covalent el-ph coupling, which is evidenced by a rather small change of the O-site energy (see Figs. 2b and 2e).

Now we turn to discuss the effects of lattice displacements on the magnetic properties. Previous studies have suggested that the quasiparticles of the low-energy excitations in an antiferromagnet can be well described by the doped hole or electron dressed by local antiferromagnetic spin fluctuations (spin-bag picture). Therefore, it is important to understand the effects of lattice displacements on local spin correlations in order to shed light on the single-particle excitation as measured in ARPES. Here, we are interested in the spin correlations of the next-nearest-neighbor (nnn) Cu-Cu sites, which are defined as

$$S(i,j) = \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle,$$

with the spin operator $\mathbf{S}_i = \sum_{\alpha\beta} d^{\alpha}_i \sigma_{\alpha\beta} d_{i\beta}$.

Figure 4 shows the nnn Cu-Cu spin correlations as a function of $u$. One can clearly see that for both hole- and electron-doped cases, the el-ph couplings have significant influences on the nnn Cu-Cu spin correlations: while the spin correlations in the direction parallel to the lattice displacement have little change, the spin correlations in the direction perpendicular to the lattice displacement increase or decrease alternatively depending on both the Cu sites and the doping. In particular, there exists a crucial difference between the hole- and electron-doped cases. In the hole-doped case, the spin correlations increase for the Cu sites with smaller charge density, and decrease for the Cu sites with larger charge density, while in the electron-doped case, this behavior is reversed.
single occupancy at the site shows the normalized spin correlations as a function of differences of the spin-phonon coupling in p- and n-type. On the other hand, we observe a rather weak effect in strong response of spin degree of freedom to phonons. From Figs. 5a and 5b, it is clear that in the hole-doped case, both ionic and covalent el-ph couplings induce a strong response of spin correlations defined as

$$S^\mu(i,j) = \frac{\langle S_i \cdot S_j \rangle}{P_{si} \cdot P_{sj}}.$$  

where $P_{si} = \langle n_i - 2n_{i\uparrow} n_{i\downarrow} \rangle$ denotes the probability of single occupancy at the site $i$. Eq. (8), thus measures the spin correlations normalized to the spin density. Figure 5 shows the normalized spin correlations as a function of $u$. From Figs. 5a and 5b, it is clear that in the hole-doped case, both ionic and covalent el-ph couplings induce a strong response of spin degree of freedom to phonons. On the other hand, we observe a rather weak effect in the electron-doped case, which is shown in Figs. 5c and 5d.

The above results indicate that there exist dramatic differences of the spin-phonon coupling in p- and n-type high-$T_c$ cuprates: the coupling of spin with lattice displacements is much stronger in the p-type materials than in the n-type materials. This could give an explanation why ARPES detected a strong mass renormalization in the p-type cuprates, while it is absent in the n-type cuprates. The reason is that within the spin-bag picture, the quasiparticle property is mainly determined by local antiferromagnetic spin fluctuations and a strong change of local spin correlations can dramatically affect the motion of doped charge carriers in the background of antiferromagnet. We argue that if the similar difference in spin-phonon couplings occurs for other phonon modes at small and large momenta, we then expect that in the n-type superconductors, the phonon contributions to electron-electron pairing and electronic transport should be much smaller than those in the p-type materials.

Summarizing, we have presented QMC studies of the effects of the half-breathing $O(\pi, 0)$ phonon mode on the ground-state properties of the 2D three-band Peierls-Hubbard model. We found that in the physically interesting parameter regime, the el-ph couplings have similar effects on the ground-state energies and charge fluctuations in both hole- and electron-doped systems. This suggests that the strong softening of the half-breathing $O(\pi, 0)$ phonon mode in both p- and n-types materials can be attributed to the the strong interactions of lattice distortions with local metallic charge fluctuations. In addition, our results show that the spin-phonon coupling is much stronger in the p-type cuprates than in the n-type cuprates. This difference in the spin-phonon coupling could provide an explanation for the difference of mass renormalization observed in ARPES experiments.

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1. A. Lanzara, P. V. Bogdanov, X. J. Zhou, S. A. Keller, D. L. Feng, E. D. Lu, T. Yoshida, H. Eisaki, A. Fujimori, K. Kishio, J.-I. Shimoyama, T. Noda, S. Uchida, Z. Hussain, and Z.-X. Shen, Nature 412, 510 (2001).
2. Z.-X. Shen, A. Lanzara, S. Ishihara, and N. Nagaosa, Phil. Mag. B 82, 1349 (2002).
3. N. P. Armitage, D. H. Lu, C. Kim, A. Damascelli, K. M. Shen, F. Ronning, D. L. Feng, P. Bogdanov, X. J. Zhou, W. L. Wang, Z. Hussain, P. K. Mang, N. Kaneko, M. Greven, Y. Onose, Y. Taguchi, Y. Tokura, and Z.-X. Shen, Phys. Rev. B 68, 064517 (2003).
4. R. J. McQueeney, Y. Petrov, T. Egami, M. Yethiraj, G. Shirane, and Y. Endoh, Phys. Rev. Lett. 82, 628 (1999); R. J. McQueeney, J. L. Sarrao, P. G. Pagliuso, P. W. Stephens, and R. Osborn, Phys. Rev. Lett. 87, 077001 (2001).
5. L. Pintschovius and M. Braden, Phys. Rev. B 60, R15039 (1999).
6. H. J. Kang, Pengcheng Dai, D. Mandrus, R. Jin, H. A. Mook, D. T. Adroja, S. M. Bennington, S.-H. Lee, and J. W. Lynn, Phys. Rev. B 66, 064506 (2002).
7. M. d’Astuto, P. K. Mang, P. Giura, A. Shukla, P. Ghigna, A. Mirone, M. Braden, M. Greven, M. Krisch, and F. Sette, Phys. Rev. Lett. 88, 167002 (2002).
8. K. Yonemitsu, A. R. Bishop, and J. Lorenzana, Phys. Rev. Lett. 69, 965 (1992).
9. Z. G. Yu, J. Zang, J. T. Gammel, and A. R. Bishop, Phys. Rev. B 57, R3241 (1998).
10. Shiwei Zhang, J. Carlson, and J. E. Gubernatis, Phys. Rev. Lett. 74, 3652 (1995); Phys. Rev. B 55, 7464 (1997).
11. J. Carlson, J. E. Gubernatis, G. Ortiz, and Shiwei Zhang, Phys. Rev. B 59, 12788 (1999).
12 S. Ishihara, T. Egami, and M. Tachiki, Phys. Rev. B 53, 3163 (1997).
13 S. Ishihara, T. Egami, and M. Tachiki, Phys. Rev. B 49, 8944 (1994).

14 J. R. Schrieffer, X.-G. Wen, and S.-C. Zhang, Phys. Rev. Lett. 60, 944 (1988).
15 R. Eder, and Y. Ohta, Phys. Rev. B 50, 10043 (1994).