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Author(s): J. Mustre de Leon, R. deCoss, A. Rubio-Ponce*
A.R. Bishop
S. A. Trugman

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Polaronic Signatures in Phonon Isotopic Shifts

J. Mustre de Leon, R. de Coss, A. Rubio-Ponce

Centro de Investigación y de Estudios Avanzados del IPN.

Merida, Yucatan 97310, Mexico

A. R. Bishop and S. A. Trugman

Los Alamos National Laboratory, Los Alamos, New Mexico 87545, USA

— DRAFT —

Abstract

The effect of O(16) by O(18) isotopic substitution in the excitation spectrum of a model electron-phonon Hamiltonian, previously used to describe the dynamics of the O(4)-Cu(1)-O(4) cluster in $YBa_2Cu_3O_7$, is presented. This model includes electronic correlations and electron-phonon interactions, exhibiting the presence of polaron tunelling. The calculated isotopic shifts of phonon excitations differ from those found using harmonic or anharmonic potentials, and are consistent results of optical measurements of c-axis phonons. The isotopic substitution changes the dynamics of polaron tunelling and produces a change in the local structure.

Several experiments in high temperature superconductors have shown the presence of polaronic carriers, in possible coexistence with Fermi-like quasiparticles. Although there is no agreement about the detailed role that such polarons play in high-Tc superconductors, there are precise signatures of the presence of these carriers. For example in microscopic properties, like the evidence a second gap independent of temperature[1] or anomalous isotopic coefficients in Tc,[2] and microscopic aspects, e.g., local lattice distortions.[3–6]

The isotope effect on Tc has been one of the more controversial results on high-Tc superconductors. This originates in part in experimental complications related to its determination, like dependence of Tc with exact stoichiometry, which can vary under isotopic substitution,[11,12] the existence of microscopic phase separation,[12] and the common happening of incomplete isotopic substitution.[11] Also, from a theoretical point of view the interpretation of the isotopic coefficient of Tc has usually relied on the assumption of coupling between quasi-electrons and harmonic phonons, which yields to BCS-like values for the isotopic shift on Tc. However, recent calculations which assume the presence of polaronic carriers[2] have shown that the isotope coefficient can differ substantially from that obtained in BCS, in consistency with experimental observations.[7] Another important aspect, which has gained recent atention, is the isotopic shift of T_s, the temperature for structural transitions, and whether this is related to the existence of polarons.[7,17] We note however, that this determination is also complicated, e.g., there are indications that while the temperature at which the average structure exhibits a change does not show an important variation with isotopic substitution,[7] the temperature at which the local stucture does change exhibits a large change under isotopic substitution.[17]

A measurement which can also show polaronic effects, put forward by isotopic substitution, is the shift of the optical excitation spectrum. We concentrate in c—axis excitations of $YBa_2Cu_3O_7$, related to the dynamics of the axial oxygen, O(4), in the infrared regime corresponding to phonon excitations. Our motivation lies in studies of the local structure which have found an anomalous non-Gaussian distribution for the axial oxygen in $YBa_2Cu_3O_7$,[4] and the existence of two axial oxygen positions in Tl-based high-Tc materials.[6] Also, optical

studies in $YBa_2Cu_3O_{7-x}$ [15] have shown that under oxygen substitution the "505 cm^{-1}" Raman active mode exhibits and isotope shift smaller than that obtained assuming a harmonic lattice dynamics.[15]

In this work we analyze the effect of isotopic shift in the lowest energy excitations of a model which consists of a three-site Hubbard Hamiltonian with Holstein hole-lattice interaction.[13] This specific model has been introduced elsewhere [13] to explain some of the novel features of the axial oxygen [O(4)] and the chain copper [Cu(1)] cluster - arranged as O(4)-Cu(1)-O(4) in $YBa_2Cu_3O_7$. We divide the Hamiltonian into an electronic part, a phononic part and an electron-phonon interaction part:

$$H = H_{\rm el} + H_{\rm ph} + H_{\rm el-ph}. \tag{1}$$

The electronic part is given by an extended Hubbard model on a three-site cluster:

$$H_{\rm el} = \sum_a \epsilon_a n_a + \sum_{ab,\sigma} t_{ab}(c^\dagger_{a\sigma} c_{b\sigma} + h.c.) + U \sum_a n_{a\uparrow} n_{a\downarrow}. \tag{2}$$

Here, $c^\dagger_{a\sigma}$ creates a hole of spin σ at site a, $n_{a\sigma} = c^\dagger_{a\sigma} c_{a\sigma}$, and $n_a = \sum_\sigma n_{a\sigma}$. Indices $a = 1, 3$ denote the axial O(4) sites and $a = 2$ is the chain Cu(1) site. The hopping matrix element is $t_{ab} = t$ between the O(4) and Cu(1) sites The site energies are parametrized by the parameter ϵ_o: $\epsilon_{1,3} = \epsilon_o$ and $\epsilon_2 = -\epsilon_o$.

For the vibrational part we consider two harmonic phonon modes corresponding to displacements in the z-direction. One mode is Raman active and the antisymmetric mode is infrared active. These are described by boson operators a_R and a_{IR} with bare frequencies ω_R and ω_{IR}. respectively. In terms of these operators. the phonon part of the Hamiltonian is

$$H_{\rm ph} = \hbar\omega_{IR} a^\dagger_{IR} a_{IR} + \hbar\omega_R a^\dagger_R a_R. \tag{3}$$

Finally, we consider a molecular-crystal type interaction between electronic and phonon degrees of freedom:

$$H_{\rm el-ph} = \lambda_{IR}(a_{IR} + a^\dagger_{IR})(n_3 n_1) + \lambda_R(a_R + a^\dagger_R)(n_1 + n_3 - s_o). \tag{4}$$

where λ_{IR} and λ_R are the respective coupling constants. The parameter s_o is chosen to avoid any artificial shrinkage of the cluster, thus allowing the use of a reduced basis set.

We assume that the the ionic charges of Cu(1) and O(4) are +1 and −2. respectively, for the closed shell configurations with no holes present. The parameters in H_{el} are taken as $\epsilon_o = 0.307$ eV, $t = -0.634$ eV, $t' = t/10$, and $U = 4.44$ eV, which are representative values for $YBa_2Cu_3O_7$, guided by local density calculations [7] For bare phonon energies, we choose $\hbar\omega_{IR} = 600$ cm^{-1} and $\hbar\omega_R = 500$ cm^{-1}, and fix $s_0 = 1.17$. We assume two holes within the O(4)-Cu(1)-O(4) cluster. In the $YBa_2Cu_3O_7$ ground-state, one hole is predominantly located at the Cu(1) site while the other fluctuates between the O(4) sites, in agreement with core-level x-ray-absorption measurements [7] implying excess holes located in the O(4) $2p_z$ orbitals.[8]

The physics of the above model was solved in Ref. [13] by exact diagonalization. For sufficiently large values of λ_{IR} a new length scale, $\delta\ell$, associated with a dynamical double-well structure in the infrared coordinate appears. The motion of the lattice is strongly correlated with the hole motion, corresponding to polaron formation, and a new time scale describing polaron tunneling is generated , $\hbar\omega_T$, defined as the energy difference between the ground state and the first excited state.

Using this model we calculated the change in the excitation frequencies as we vary the mass of the oxygen ion (sites 1 and 3) from O(16) to O(18). We note the explicit dependence on the oxygen mass of the bare phonon frequencies and the electron-lattice coupling Hamiltonian. We consider the relative shift:$[\omega(O_{16}) - \omega(O_{18})/\omega(O_{16})]$ for the low-lying excitations. In the intermediate coupling regime, the isotopic shift for the first excitation (polaron tunneling) is negative, 4.3%. The isotopic shift for higher excitations differs from that found using harmonic or anharmonic potentials, see Table I. In this table we also present experimentally measured values.[14–16] From this comparison we conclude that the isotopic shifts calculated with the proposed model, differ from the harmonic values and are consistent with the experimentally observed values. We note however, that other factors could also explain the discrepancies observed between the experimental values and lattice dynamics calcula-

tions which assume harmonic potentials, - e.g., uncertainties in the determination of normal mode eigenvectors could change the values obtained in lattice dynamics calculations. Under isotopic substitution, in this polaronic regime, we obtained a change in the two site structure of $\sim$.01 Å. We note that a change in local lattice distortions with isotopic substitution in $LaCuO_{4+\delta}$ has been recently reported.[17] This change was interpreted as a shift in T_s under isotpic substitution. This observation is also consistent with the change in dynamics of the poalron tunneling described above.

We have studied the effect of isotopic substitution in a model interacting electron-lattice system, which exhibits polaron tunneling. We find a change in the polaron tunneling dynamics, and a change in the local structure. We also find isotopic shifts in phonon excitations which differ from those derived using harmonic or anharmonic rigid potentials. These results are consistent with recent experimental observations, giving added evidence for the presence of polaronic effects in high-Tc superconductors. As dicussed in Ref. 9, the coupling of these carriers with quasi-electrons has important implications for properties of these materials and might be a key element in understanding the origin of high-Tc superconductivity. This work was supported by CONACyT, Mexico, and U.S. Department of Energy.

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TABLES

TABLE I. Optical shift frequencies in $YBa_2Cu_3O_7$. Experimental and calculated values of $\Delta\omega/\omega(\%)$ for the Raman and Infrared modes.

Mode	Exp.	Shell model	harmonic	pol. mod.
Raman active	4.70± 0.4[a]	5.60[a]	5.77	4.66
Infrared active	3.60 ± 0.4[b]	4.33[c]	3.70	3.34

[a] Reference [15].

[b] Reference [14].

[c] Reference [16].