Microstrain and polaronic correlation in a model system

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Abstract. We have studied the effect that variations of the local microstrain have on the polaronic correlation function obtained from the exact diagonalization of a simple model Hamiltonian. Although the correlation function exhibits a monotonic behavior as a function of the hole hopping parameter, and thus microstrain, its isotopic shift exhibits a maximum for specific values of $t$. This result suggests that if there is a coupling between polaronic objects and free carriers, coexisting in high-$T_c$ cuprate superconductors, then this will manifest not only in the superconducting transition, but also on the properties directly related to the polaron formation in these materials.

1. Introduction

It has been proposed that the electronic properties in high temperature superconductor cuprates differ from those of a Fermi liquid due to the proximity of these systems to a quantum phase transition. [1] A quantum phase transition is a zero temperature phase transition that is controlled by a parameter of the Hamiltonian that defines the dynamics of the system in question. In the vicinity of such transition fluctuations take the system between two different ground states, leading to a spatially inhomogeneous system.

In the case of high temperature superconductors this quantum phase transition could be related to the disappearance of an antiferromagnetic ground state as doping is increased [2,3]. However, the maximum on $T_c$ is observed for doping levels ~ 0.15, while the disappearance of the AF order occurs at much lower doping levels ~ 0.06 [4]. Thus not supporting this hypothesis. More recently the observation of local lattice distortions, related to the presence of pseudo Jahn-Teller polarons, suggested a quantum phase transition between a polaronic ordered state and a free carrier ground state [5,6].

Bianconi and co-workers have proposed that the variable that would control the proximity to a quantum critical point would be the tensile microstrain, $e$, and that together with doping defines a such quantum critical point (QCP) around a maximum $T_c$ [5,6]. Two nearly degenerate ground states fluctuate around this QCP; an inhomogeneous phase of polarons arranged in stripe-like structures and free charge carriers. The tensile microstrain, $e$, originates in the mismatch between the $CuO_2$ layer and the rock-salt layer in cuprate perovskites. This mismatch depends on the variation in the sum of radius of the metal-ion in the rock-salt layer and the radius of $O^2-$ in the $CuO_2$ plane. This leads to variations of the $Cu-O$ bond length and thus modifies transport and other properties from those of a system that does not present a microstrain. In fact this has been postulated as a possible explanation for reports of increased $T_c$ of films grown under tensile strain.
It has also been proposed that the enhanced $T_c$ of the cuprates is related to an exchange resonant effect between the free carriers in the Fermi liquid component and the charges that are part of the polaron. Consequently, there is an optimal value of the microstrain that will maximize this exchange.

For these reasons, we decided to investigate the effect of microstrain variations on a simple model Hamiltonian, which exhibits polaronic behavior for moderate and strong values of the electron-lattice coupling [7-9]. In this case, following the premise of Ref. [5] we assume that the main effect of the microstrain variation is the modification of the nearest-neighbor hole hoping term, $t$. We specifically concentrate on the effect that the change in $t$, has on the charge-distortion correlation function that defines the polaronic appearance on the used model Hamiltonian.

### 2. The electron-phonon Hamiltonian

The model Hamiltonian we have used describes the dynamics of two holes in a three-site cluster. It is the smallest cluster which can show explicitly polaron tunneling. Although this model has been used before identifying the particular cluster with the O(4)-Cu-O(4) cluster in $YBa_2Cu_3O_7$ [7-9], it can describe in general the motion of two holes in similar regions separated by a domain wall, using the appropriate parameters that enter in the Hamiltonian. For the sake of completeness we present the elements, which describe the model Hamiltonian here,

$$ H = H_e + H_{ph} + H_{e-ph} $$

The electronic part of the Hamiltonian takes the form of a single band Hubbard model,

$$ H_e = \sum_{\sigma, i=1}^{\beta} e_n \sigma + U \sum_{\sigma} n_{\sigma} n_{\bar{\sigma}} + V \sum \langle n \rangle \sigma + t \sum_{\sigma} (c^\dagger_{\sigma \alpha} c_{\sigma \alpha} + c^\dagger_{\sigma \alpha} c_{\bar{\sigma} \beta} + c^\dagger_{\bar{\sigma} \beta} c_{\sigma \alpha} + H.c). $$  \hspace{1cm} (1)

$U$ is the onsite Coulomb interaction, $t$ is the hopping energy between adjacent sites, and $\varepsilon$ the site charge energy in an atomic orbit. The superscripts $i=1,3$ are for two oxygen sites and $i=2$ for the only copper site. The site energies are parameterized with, $\varepsilon_1 = \varepsilon_3 = -\varepsilon_2 = \varepsilon_0$. The number operator is defined by $n_{\sigma} = c^\dagger_{\sigma \alpha} c_{\sigma \alpha}$, where the operators $c^\dagger_{\sigma \alpha}$ creates a hole with spin, $\sigma$, $c_{\sigma \alpha}$ destroys it and $n_{\sigma} = \sum_{\sigma} n_{\sigma} \sigma$.

For the phononic part of the Hamiltonian, we consider the symmetric and anti-symmetric modes for a three-site cluster, with bare frequency $\omega_0^S$, $\omega_0^A$. In terms of bosonic creation and annihilation operators $b_j^{S}$ and $b_j^{A}$, the phononic term is written down as,

$$ H_{ph} = \hbar \sum_j \omega_j b_j^{S} b_j^{S} + \hbar \omega_j^{A} b_j^{A} b_j^{A}. $$

The relation between the normal coordinates and the bosonic operators is

$$ u_j = \left( \frac{\hbar}{2m_0 \omega_0^j} \right)^{1/2} (b_j^{S} + b_j^{A}). $$

The third term defines the electron-lattice interaction. We use an interaction that couples the hole motion with the phononic modes

$$ H_e^{\text{ionic}} = -\Lambda_S u_S \sum_{\sigma} (n_{1\sigma} - 2n_{2\sigma} + n_{3\sigma}) - \Lambda_A u_A \sum_{\sigma} (n_{3\sigma} - n_{1\sigma}). $$

Here $\Lambda_S = (\hbar/2m_0 \omega_0^S)^{1/2}$ $\Lambda_S$ couples hole motion with the symmetric mode and $\Lambda_A = (\hbar/2m_0 \omega_0^A)^{1/2} \Lambda_S$ couples with the antisymmetric mode. For the O(4)-Cu(1)-O(4) chain in $YBa_2Cu_3O_7$, the Raman mode is set as the symmetric mode ($j=R$) and the infrared mode to the antisymmetric mode. We identify the asymmetric mode with the half breathing mode in the CuO$_4$
cluster for the CuO plane in La₂CuO₄ and the symmetric mode with the breathing mode. The parameters used in (1) are taken from [9], \( U = 7.0 \text{ eV} \), \( \varepsilon_0 = 0.5 \text{ eV} \), \( t_0 = -0.5 \text{ eV} \). As explained above later on \( t \) will be used as a variable, related to the local microstrain. The bare phonon modes are fixed to values found experimentally in optical and inelastic neutron scattering experiments: \( \omega_\Lambda^0 = \omega_K^0 = 500 \text{ cm}^{-1} \) and \( \omega_\Lambda^0 = \omega_K^0 = 612.4 \text{ cm}^{-1} \) for YBa₂Cu₃O₇.

Since only the coupling between the asymmetric mode and the charge motion leads to a measurable lattice distortion, with two Cu-O bond lengths [7] we only consider the effect of the variation in the electron-phonon coupling constant with the antisymmetric mode. For intermediate values of the electron-phonon coupling a polaronic dynamics develops, i.e. same time or energy scale in the ionic and hole motion. We fix \( \lambda_s = 0.1 \text{ eV} \). It is important to note that for intermediate electron-phonon coupling values \( \lambda = 0.13 \text{ eV} \), a distortion in the Cu-O bond develops with a split Cu-O distance as those found in EXAFS experiments, yielding to a nonzero value of the average \( u_{IR} \approx \delta [7,8] \). Another signature of the polaronic behavior present for these intermediate values of the electron-phonon coupling is the negative value of the energy shift under \(^{16}\text{O}\) by \(^{18}\text{O}\) substitution [10].

3. Modelling microstrain in the three-site model.

We introduce microstrain variation effects through the variation of the hopping term. In order to model the stretch effect of the Cu-O bond-length we use values of \( t \) lower than \( t_0 \) and to model the compression of a bond length we use values higher than \( t_0 = 0.5 \text{ eV} \). This assumption is based on the fact that the hopping reflects the overlapping of atomic orbitals and therefore it has an exponential dependence with the bond length. We note however, that phonon modes and other electronic parameters might also change, however the hopping parameter, \( t \), is more sensitive to bond length changes due to its exponential dependence.

It is important to study the effect of microstrain for different values of the electron-phonon coupling. We choose \( \lambda = 0.10 \text{ eV} \) as the coupling for the onset of the polaron regime and two other couplings, \( \lambda = 0.13 \text{ eV} \), for a description of slower polaron tunnelling state, and \( \lambda = 0.05 \text{ eV} \) for a system in which polaronic behavior has not appeared yet. All these cases correspond to values of \( t = t_0 = 0.5 \text{ eV} \).

![Figure 1. Polaronic correlation function C as a function of t.](image)
We define the polaronic correlation function $C = \langle u_{IR}(n_3-n_1) \rangle / \delta$, where the average value is taken over the ground state of the Hamiltonian, and $\delta$ corresponds to the average value of the infrared coordinate $u_{IR}$, as described in the previous section. We note that if the charge motion is not correlated with the motion of the ions $C \rightarrow 0$, and as the motion becomes correlated $C$ is expected to grow. Other definitions of correlation show the same qualitative behavior [11].

Figure 1 shows the results of the effect of microstrain variation (or $t$) on the correlation function. We can see that as $t$ increases $C$ decreases, and the value of $C$ is larger for larger values of the electron-lattice coupling constant. These results indicate that for a given electron-lattice coupling constant the effect of faster bare hole hopping tends to destroy the polaronic state. These results support the intuitive expectation that as the charge hopping increases a stronger electron-phonon coupling is needed to correlate the charge and ionic dynamics in order to form a polaronic state.

4. Isotopic shifts of the polaronic correlation function
The results presented in figure 1 show a monotonic behavior of the correlation function as the bare hole hopping is increased. This increase in $t$ would correspond to an increase in the tensile stress $e$. Hence, it does support the intuitive expectation that as the microstrain is increased, the Cu-O bond lengths become smaller yielding a faster hole hopping rate that destroys the correlation between ionic and hole motion. We note that the parameters in the Hamiltonian, specifically the electron-lattice coupling constant, can be chosen to reproduce the experimental observation of local lattice distortions with Cu-O bond length differences of the order of 0.1 Å [7,8]. In this case the dynamical time scale of the polaronic tunneling becomes of the order of 10-20 meV, smaller than typical phonon excitations. Also a negative isotopic shift of the polaronic tunneling frequency is observed under $^{16}$O by $^{18}$O substitution, yielding a clear indication that the observed local lattice distortions are indeed related to the presence of polarons in the system [10].

However, the polaronic correlation function, $C$, in absence of an explicit interaction with free carriers does not show a particular dynamical time or length scale, that would reflect a resonant exchange, even for values of $t$ that reproduce distortions consistent with the experimental observation of local lattice distortions of the order of 0.1 Å.

Nevertheless, in this simplified model, the parameters entering in the Hamiltonian are chosen in such a way that the generated polaronic behavior reproduces the experimentally observed local lattice distortions. Consequently the model parameters represent effective parameters that should implicitly reflect the presence of all carriers in the system, including free charge carriers. Therefore we might expect that some other property that related to the polaronic behavior might actually exhibit a characteristic scale, suggesting the presence of a resonant exchange.

Given this expectation we calculated the change of the correlation function $C$ under $^{16}$O by $^{18}$O isotopic substitution. As mentioned before another signal of the onset of polaronic behavior is the negative isotope effect of the excitation energy corresponding to polaronic tunnelling. In this calculation the isotopic substitution $^{16}$O by $^{18}$O modifies the bare phonon frequencies yielding to new phononic frequencies, $\omega_1^{16}$ and $\omega_1^{18}$, given by the scaling of normal modes in a three-body-chain system coupled by an elastic medium. Because the electron-phonon coupling constant is mass dependent, it also changes by the isotopic substitution. Figure 2 shows the results for the calculation of the correlation function for $^{16}$O by $^{18}$O and its normalized percentage change, i.e., $100 \times (C_{^{16}O} - C_{^{18}O}) / C_{^{16}O}$, for an electron-lattice coupling constant, $\lambda = 0.13$ eV.

Unlike the polaronic correlation function, $C$, the behavior of the isotopic change of $C$ is non monotonic (see Fig. 2). It reaches a maximum as a function of $t$ for $t \sim t_0 = 0.5$ eV that is the characteristic value for which the lattice distortions of the order $\sim 0.1$ Å are reproduced. There seems to be an optimal combination of values of charge hopping and electron-phonon coupling that leads to a maximum in the isotopic shift of the polaronic correlation function. In order to evaluate the generality of the observed behavior we calculated the isotopic shift of $C$, for different values of the electron-lattice coupling constant $\lambda$. We present the results in figure 3.
In this case we find that for values of $\lambda \geq 0.10$ eV, the regime in which polaronic behavior is present, there is always a maximum on the isotopic shift of $C$. We note that in these calculations all other parameters entering in the Hamiltonian, i.e., $\varepsilon, U$, were kept fixed.

![Figure 2](image2.png)

**Figure 2.** Isotopic change of the polaronic correlation function, $C$.

![Figure 3](image3.png)

**Figure 3.** Isotopic change of the polaronic correlation, $C$, for different values of the electron-lattice coupling constant.

5. **Summary and Conclusions**

We have studied the effect that variations of the local microstrain have on the polaronic correlation function obtained from the exact diagonalization of a simple model Hamiltonian. Although the correlation function exhibits a monotonic behavior as a function of the hole hopping parameter, and thus microstrain, its isotopic shift exhibits a maximum for specific values of $t$. This observation is similar to the experimental observation of an optimal $Cu-O$ distance (and thus a $t$) for the highest $T_c$. 
This result suggests that if there is a coupling between polaronic objects and free carriers coexisting in high-$T_c$ cuprate superconductors, as that proposed in Refs. [5,6,12], then this will manifest not only in the superconducting transition, but also on the properties directly related to the polaron formation in these materials. It would be important to check whether in other materials that exhibit presence of polarons, e.g., manganites [13] and nickelates[14], the parameters that will lead to the experimentally observed lattice distortions will lead to the behavior of the polaronic correlation function found in this work.

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