A variational approach to the optimized phonon technique for electron-phonon problems

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An optimized phonon approach for the numerical diagonalization of interacting electron-phonon systems is proposed. The variational method is based on an expansion in coherent states that leads to a dramatic truncation in the phonon space. The reliability of the approach is demonstrated for the extended Holstein model showing that different types of lattice distortions are present at intermediate electron-phonon couplings as observed in strongly correlated systems. The connection with the density matrix renormalization group is discussed.

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The interaction of electrons with local lattice deformations plays an important role in many different materials leading to a number of effects that range from collosal magnetoresistance in manganites [1] to superconductivity in fullerenes [2] from pseudo-gap in cuprates [3] to Peierls instability in quasi-one-dimensional materials, [4] only to mention a few. Although the theoretical study of the electron-phonon (e-ph) interaction has attracted constantly the interest of the scientific community, several aspects are still not fully understood and challenging. Actually, analytical solutions of the e-ph problem are available only in weak and strong coupling asymptotic regimes, even for the simplest molecular crystal model introduced by Holstein. [5] Some insight in the problem for intermediate coupling comes from the dynamical field approach [6] that provides exact results in the infinite dimension limit and variational methods. [7] In order to achieve a complete understanding of the e-ph interaction, numerical techniques as exact diagonalization and Quantum Monte Carlo have been exploited. In both cases the specific problem posed by the e-ph interaction is related to the presence of phonons that require an infinite dimensional Hilbert space. As a consequence, for instance, the direct application of Lanczos exact diagonalization has been limited to very small lattices and needs a somehow arbitrary truncation in the phonon number. In this context the introduction of an optimized phonon approach based on the analysis of the density matrix [8, 9] has produced an important improvement. The idea beyond this approach is to take advantage of the knowledge of the largest eigenvalues and eigenvectors of the site density matrix to select the phonon linear combinations that, at the best, can describe the system: the so-called optimized phonon basis (OPB). Unfortunately, the density matrix of the target states is not available a priori; it must be calculated in a self-consistent way together with the OPB. To this aim different strategies have been discussed. In this report we wish to introduce a variational technique, based on an expansion in coherent states, that very much simplifies the selection of an optimized phonon basis and that does not require any truncation in the number of phonons. The method is able to provide accurate results for any e-ph coupling regime and for any value of the adiabatic ratio. As we will show in the case of the Holstein model, the proposed expansion provides states surprisingly close to the eigenvectors of the site density matrix corresponding to the highest probabilities and allows us to clarify some questions of the e-ph interaction that have been debated recently. In particular, the role of quantum lattice fluctuations in the Holstein model at half-filling and in the clustered phase is discussed emphasizing the coexistence of very different lattice deformations that all contribute to the ground state.

The model and the proposed optimized phonon basis.

In the framework of e-ph systems the Holstein model has been very much studied. Here we discuss a simple generalization that includes nearest neighbor e-ph interaction terms. Furthermore, we consider spinless fermions in order to simulate at a very rough level [10] a strong on-site electron repulsion. In the 1D case the Hamiltonian is

$$H = -t \sum_i \left( c_i^\dagger c_{i+1} + c_{i+1}^\dagger c_i + h.c. \right) + \omega \sum_i b_i^\dagger b_i + g \omega \sum_i c_i^\dagger c_i \left( b_i^\dagger + b_i \right) + \varepsilon \sum_{\delta = \pm 1} \left( b_{i+\delta}^\dagger + b_{i+\delta} \right)$$

(1)

where $c_i^\dagger$ ($c_i$) is the site electron creation (annihilation) operator and $b_i^\dagger$ ($b_i$) creates (annihilates) a phonon in the site $i$. The phonon frequency is $\omega$, the electron hopping between nearest neighbors is controlled by $t$ while $g$ and $g \omega$ describe the strength of the on-site and nearest neighbor e-ph interactions, respectively. We assume $\hbar = 1$.

The "natural" basis in which we can describe Hamiltonian (1) is given by

$$|\nu, \mu\rangle = \prod_i |\nu_i\rangle |\mu_i\rangle,$$

(2)

where $i$ run over the lattice sites, $\nu_i$ is the electron state label that, for spinless electrons, can assume only two values and $\mu_i$ labels the infinite phonon states on-site.
Our aim is to achieve a very satisfying estimation of the ground state of $|\mu_i\rangle$ by using only a finite number of states. The first step is to replace the phonon states $|\mu_i\rangle$ at site $i$ with coherent states (CS)

$$|h, i\rangle = e^{g(h_i - b_i^\dagger)} |0\rangle^{(ph)}_i,$$

where $|0\rangle^{(ph)}_i$ is the phonon vacuum at the site $i$. This substitution does not introduce any truncation in the Hilbert space since, varying $h$ in the complex plane, the local basis is over-complete. The second step is to choose a finite number $M$ of CS and the corresponding values $h_\alpha$ ($\alpha = 1, ..., M$) that we have supposed not to depend on the site. The most efficient way to proceed is to fix the number of CS arbitrarily and choose the $h_\alpha$ variationally calculating the ground state energy and minimizing it. In this way we obtain the "best" subspace where to find the ground state. The method has the advantage to allow a systematic improvement of the approximation adding more and more CS and provides a quantitative estimation of the error introduced by a specific truncation. Of course, the number of CS per site cannot grow as we wish since the size of the matrix to be transformed depends neither on site $i$ nor on the specific CS, on physical ground, the lattice deformation of site $i$ is expected to depend very much on whether or not the site is occupied by an electron. This can be done acting with the operator $S$ of the usual Lang-Firsov transformation on each CS belonging to the chosen basis:

$$S = e^{g(\langle n_i\rangle + f(n_i - \langle n_i\rangle))(b_i - b_i^\dagger)},$$

where $f$ depends neither on site $i$ nor on the specific CS to which $S$ is applied. The value of $f$ can be estimated variationally together with the set $\{h_\alpha\}$. Technically, it is easier first to work out the action of $S$ transforming $H$ to $H' = S^\dagger HS$ and, then, to study the properties of $H'$ in the subspace spanned by both the electron and the optimized phonon basis:

$$|\{h_\alpha\}\rangle = \prod_{i=1}^N |h_\alpha, i\rangle = \prod_{i=1}^N \left[ e^{g h_\alpha_i (b_i - b_i^\dagger)} |0\rangle^{(ph)}_i \right],$$

where $h_\alpha_i \in \{h_\alpha\}$ and $\alpha = 1, ..., M$.

**Single polaron features and site density matrix.** In order to clarify the relation between the proposed approach and the optimized phonon states obtained within the density matrix method, we study a two-site one-electron system. In particular, we can construct the site density matrix calculating its eigenvalues and eigenvectors. Both these quantities can be evaluated both within our approach and by an exact diagonalization method in the "natural" phonon basis. The comparison between the two results is contained in Fig.1 where the two eigenvectors corresponding to the largest eigenvalues are plotted as a function of the phonon number for $\beta = 1$ (occupied site). It is clear that already with two CS the agreement is surprisingly good while with three CS the differences with the numerically exact results are completely negligible. The agreement is due to the fact that the exact site density matrix contains only a few relevant eigenvalues and that the corresponding eigenvectors can be approximated as linear combinations of CS. It is worthwhile noting that, roughly speaking, in the strong and intermediate coupling regime, the two most important eigenvectors correspond to strong (weak) lattice deformations and weak (strong) lattice deformations, respectively. The two contributions are both important in the so-called intermediate regime while in the weak and strong regimes only one type of lattice deformation is relevant. Also for $\beta = 0$ (empty site) the site deformation is built up through both weak and/or strong contributions; however, the method proposed is so flexible to allow different distortions for empty and occupied sites. These observations make clear why the generalized Lang-Firsov method, based on a
 Evidence of quantum fluctuations in clustered phases at intermediate coupling. Recently the formation of clusters due to e-ph interaction has been discussed in connection to experimental evidences of charge ordering in cuprates and manganites. One of the simplest models that supports cluster formation is the modified Holstein model of eq.\textsuperscript{[16]}. In the weak coupling regime the system is expected to form a Luttinger liquid, while, for \( \varepsilon \neq 0 \) the effective electron interaction, that is attractive for nearest neighbors, is responsible for the formation of electron clusters at strong coupling. As we go from strong to weak coupling, the cluster tends to break up and, in the intermediate coupling regime and for phonon and electron energy scales not well separated (\( \omega \simeq t \)), all the electron configurations contribute to the ground state without a dominant configuration (Fig.4). In this regime, that is the most interesting both from the experimental and the theoretical point of view, our calculation shows that different electron configurations

The half-filling case: anti-adiabatic regime and quantum fluctuations. It is well known that the ground state of the 1D Holstein model exhibits a phase transition from a Luttinger liquid to a charge ordering state at half-filling, but the role of the quantum fluctuations has not been fully explored. First of all we have checked that the total energy and the static structure factor calculated with our method are in excellent agreement with previous estimations based on exact diagonalization obtained by Lanczos method. In particular, in the intermediate regime (the worst case), our estimations based on two CS is 0.1% higher than the best estimation available. In order to understand the role of the quantum fluctuations in the anti-adiabatic regime (\( \omega \gg t \)) where quantum lattice effects are expected to play an important role, we have calculated \( P(X;\chi) \), the lattice displacement probability distribution function (LDPDF), at a fixed electron configuration \( \chi \). In Fig. 3 we show the results of the calculated LDPDF, relative to a specific site, associated to the most important electron configurations close to the boundary of the CO phase for \( \omega = 5t \) and \( g^2 = 3.6 \).

The full line gives the LDPDF of an occupied site in the CO electron configuration where alternating sites are occupied. As expected it is peaked at a large value of the lattice displacement (roughly \( X_0\sqrt{2M\omega} \simeq 2g \simeq 3.8 \), \( M \) being the ionic mass) since we are in the strong coupling regime. However, in Fig. 3 it is shown that there are less important electron configurations among which some contribute to the LDPDF with a peak at around \( X_0\sqrt{2M\omega} \simeq 0 \) typical of empty sites. These less important electron configurations can be viewed as "defects" with respect to the "ideal" CO configuration. These defects are associated to very different lattice displacements compared to the ones characteristic of the dominant CO configuration, but are still important in the anti-adiabatic regime. We expect that this behavior is still present in the thermodynamic limit since the electron configurations shown in Fig.3 are relevant also in the broken symmetry phase of the thermodynamic limit. We note that the strong fluctuations represent the quantum counterpart of the thermal fluctuations found in the opposite adiabatic limit within the dynamical mean field approach (DMF). Furthermore, we stress that, in the adiabatic limit, the quantum fluctuations behave in a very different way giving rise to "defects" whose lattice displacement is of the same type of the dominant CO configuration. For this reason, as shown in DMF approach, the ground state LDPDF is characterized by a single peak.

\begin{table}
\centering
\begin{tabular}{|c|c|c|c|c|}
\hline
\( g^2 \omega/t \) & \( E/t\) \textsuperscript{[15]} & 2CS & 3CS & 4CS \\
\hline
0.5 & 0.4 & -2.06130 & -2.06128 & -2.06130 & -2.06130 \\
5.0 & 0.4 & -2.72827 & -2.71365 & -2.72777 & -2.72828 \\
1.0 & 4.0 & -5.02985 & -5.02883 & -5.02984 & -5.02985 \\
\hline
\end{tabular}
\caption{The ground state energy \( E/t \) of a polaron for different regimes by using 2, 3, 4 CS per site is compared with results provided by Wellein et al.\textsuperscript{[15]}. The system size is \( L = 10 \).}
\end{table}

\begin{figure}
\centering
\includegraphics[width=0.5\textwidth]{fig2}
\caption{The ground-state energy of a polaron as a function of the coupling constant \( \lambda = g^2\omega/2t \) for different adiabatic ratios \( \omega/t \). The case \( \omega/t = 1 \) is compared with the data of Ref.\textsuperscript{[12]}.}
\end{figure}
FIG. 3: The LDPDF of the site indicated by the arrow for the most important electron configurations are plotted as a function of the lattice distortion in the anti-adiabatic region and in the charge-ordering regime. There are important electron configurations characterized by very different distortions. The lattice distortions are measured in units of $(2M\omega)^{-1/2}$.

can be associated to very different lattice deformations. This result stems out clearly from Fig.4 where we have plotted the LDPDF of two electron configurations for $\lambda = 1.2$ and $\varepsilon = 0.1$. The site occupied by the central electron in the cluster configuration (a) is characterized by a LDPDF peaked at high values of deformation while the site occupied by the central electron in configuration (b) presents much weaker deformations typical of the weak coupling regime with larger spreading. The balance between effective hopping, reduced by the cluster formation, and e-ph energy gain can allow a larger lattice deformation energy. The presence of electron configurations with very different lattice deformations that contribute to the ground state almost at same level suggests the existence of significant quantum dynamical fluctuations triggered by e-ph interactions. We think that our result, although limited to a simple 1D e-ph model, provides support to a number of experimental evidences in cuprates, manganites, where lattice distortions have been observed by infrared, neutron scattering and X-ray spectroscopy.

Conclusions. In this report we have proposed a variational approach, based on CS expansion, that is able to identify optimized phonon basis useful for studying e-ph models. The reliability of the model has been demonstrated for the extended Holstein model emphasizing the role of quantum fluctuations. In particular, it has been pointed out that very different lattice distortions coexist at intermediate e-ph coupling.

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