Optimized phonon approach for the diagonalization of electron-phonon problems

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We propose an optimized phonon approach for the numerical diagonalization of interacting electron-phonon systems combining density-matrix and Lanczos algorithms. We demonstrate the reliablity of this approach by calculating the phase diagram for bipolaron formation in the one-dimensional Holstein-Hubbard model, and the Luttinger parameters for the metallic phase of the half-filled one-dimensional Holstein model of spinless fermions.

Problems of electrons or spins interacting with lattice degrees of freedom play an important role in condensed-matter physics. To name only a few, consider for instance polaron and bipolaron formation in various transition metal oxides such as tungsten oxide or high-T_c cuprates,[1] Jahn-Teller effects in colossal magnetoresistance manganites,[2] or Peierls and spin-Peierls instabilities in quasi-one-dimensional materials.[3]

As a generic model for such systems the one-dimensional (1D) Holstein-Hubbard model,

$$\mathcal{H}=-t\sum_{i,\sigma}\,(c^{\dagger}_{i,\sigma}c_{i+1,\sigma}+\text{H.c.})+U\sum_{i}\,n_{i,\downarrow}n_{i,\uparrow}$$

$$+g\omega\sum_{i,\sigma}\,(b^{\dagger}_{i}+b_{i})c^{\dagger}_{i,\sigma}c_{i,\sigma}+\omega\sum_{i}\,b^{\dagger}_{i}b_{i}\,, \qquad (1)$$

is frequently considered, where $c^{(\dagger)}_{i,\sigma}$ and $b^{(\dagger)}_{i}$ describe fermions and bosons on a site i, respectively. In many physically relevant situations the energy scales of electrons (t,U), phonons (ω) and their interaction $(g\omega)$ are of the same order of magnitude, causing analytic methods, and especially adiabatic techniques, to fail in most of these cases. Even for numerical methods strong interactions are a demanding task, since they require some cut-off in the phonon Hilbert space. Starting with the work of White[4] in 1993, during the last years a class of algorithms became very popular, based on the use of a so-called density matrix for the reduction of large Hilbert spaces to manageable dimensions. Considerable focus has been placed on renormalization methods for one-dimensional systems in the thermodynamic limit. However, exact diagonalization of finite clusters also benefit substantially from these ideas, as we will demonstrate in the present paper.

Optimized phonon approach. First we resume the connection between density matrices and optimized basis states. Starting with an arbitrary normalized quantum state

$$|\psi\rangle=\sum_{r=0}^{D_r-1}\sum_{\nu=0}^{D_\nu-1}\,\gamma_{\nu r}|\nu\rangle|r\rangle \qquad (2)$$

expressed in terms of the basis $\{|\nu\rangle|r\rangle\}$ of the direct product space $H=H_\nu\otimes H_r$, we wish to reduce the dimension D_ν of the space H_ν by introducing a new basis,

$$|\tilde{\nu}\rangle=\sum_{\nu=0}^{D_\nu-1}\,\alpha^{\tilde{}}_{\tilde{\nu}\nu}|\nu\rangle, \qquad (3)$$

with $\tilde{\nu}=0\ldots(D_{\tilde{\nu}}-1)$ and $D_{\tilde{\nu}}<D_\nu$. We call $\{|\tilde{\nu}\rangle\}$ an optimized basis, if the projection of $|\psi\rangle$ on the corresponding subspace $\tilde{H}=H_{\tilde{\nu}}\otimes H_r\subset H$ is as close as possible to the original state. Therefore we minimize $\||\psi\rangle-|\tilde{\psi}\rangle\|^2$ with respect to the $\alpha^{\tilde{}}_{\tilde{\nu}\nu}$ under the condition $\langle\tilde{\nu}'|\tilde{\nu}\rangle=\delta_{\tilde{\nu}'\tilde{\nu}}$, where

$$|\tilde{\psi}\rangle=\sum_{r=0}^{D_r-1}\sum_{\tilde{\nu}=0}^{D_{\tilde{\nu}}-1}\sum_{\nu,\nu'=0}^{D_\nu-1}\,\alpha^{\tilde{}}_{\tilde{\nu}\nu}\alpha^{*}_{\tilde{\nu}\nu'}\gamma_{\nu' r}|\nu\rangle|r\rangle \qquad (4)$$

is the projected state. Since we find

$$\||\psi\rangle-|\tilde{\psi}\rangle\|^2=1-\sum_{r=0}^{D_r-1}\sum_{\tilde{\nu}=0}^{D_{\tilde{\nu}}-1}\sum_{\nu,\nu'=0}^{D_\nu-1}\,\alpha^{\tilde{}}_{\tilde{\nu}\nu}\gamma^{*}_{\nu r}\gamma_{\nu' r}\alpha^{*}_{\tilde{\nu}\nu'}$$

$$=1-\mathrm{Tr}(\boldsymbol{\alpha}\boldsymbol{\rho}\boldsymbol{\alpha}^{\dagger}), \qquad (5)$$

where $\boldsymbol{\rho}=\Sigma_{r=0}^{D_r-1}\gamma^{*}_{\nu r}\gamma_{\nu' r}$ is called the density matrix of the state $|\psi\rangle$, we observe immediately that the states $\{|\tilde{\nu}\rangle\}$ are optimal if they are elements of the eigenspace of $\boldsymbol{\rho}$ corresponding to its $D_{\tilde{\nu}}$ largest eigenvalues $w_{\tilde{\nu}}$.

Following Zhang *et al.*,[5] we apply these features to construct an optimized phonon basis for the eigenstates of an interacting electron/spin-phonon system. Consider a system composed of N sites, each contributing a phonon degree of freedom $|\nu_i\rangle$, $\nu_i=0\ldots\infty$, and some other (spin or electronic) states $|r_i\rangle$. Hence, the Hilbert space of the model under consideration is spanned by the basis $\{\otimes_{i=0}^{N-1}|\nu_i\rangle|r_i\rangle\}$. Of course, to numerically diagonalize a Hamiltonian operat-

ing on this space, we need to restrict ourselves to a finite-dimensional subspace. To calculate, for instance, the lowest eigenstates of the Holstein-Hubbard model (1), we could limit the phonon space spanned by $|\nu_i\rangle=(\nu_i!)^{-1/2}(b_i^\dagger)^{\nu_i}|0\rangle$ by allowing only the states $\nu_i<D_i$. Most simply we can choose $D_i=M\ \ \forall\ \ i$ yielding $D_{\rm ph}=M^N$ for the dimension of the total phonon space. However, if we think of the states $\{\otimes_{i=0}^{N-1}|\nu_i\rangle\}$ as eigenstates of the Hamiltonian $\mathcal{H}_{\rm ph}=\omega\Sigma_{i=0}^{N-1}b_i^\dagger b_i$, it is more suitable for most problems to choose an energy cut-off instead. Thus we used the condition $\Sigma_{i=0}^{N-1}\nu_i<M$, leading to $D_{\rm ph}=\binom{N+M-1}{N}$, for most of our previous numerical work (see, e.g., Ref. 6). For weakly interacting systems already a small number M of phonon states is sufficient to reach very good convergence for ground states and low-lying excitations. However, with increasing coupling strength most systems require a large number of the above ''bare'' phonons, thus exceeding capacities of even large supercomputers. In some cases one can avoid these problems by choosing an appropriate unitary transformation of the Hamiltonian, but in general it is desirable to find an optimized basis automatically.

Within the present density-matrix algorithm[5] for the construction of an optimal phonon basis the phonon subsystem is considered as a product of one ''large'' and a number of ''small'' sites. Each site except the large one uses the same optimized basis $\{|\mu_i\rangle\}=\{|\tilde{\nu}\rangle\}$ with $\tilde{\nu}=0\ldots(m-1)$, while the basis of the large site consists of the states $\{|\tilde{\nu}\rangle\}$ plus some bare states $\{|\nu\rangle\}$, $\{|\mu_0\rangle\}=\mathrm{ON}(\{|\tilde{\nu}\rangle\}\cup\{|\nu\rangle\})$, where ON(...) denotes orthonormalization. After a first initialization the optimized states are improved iteratively through the following steps

(1) calculating the requested eigenstate $|\psi\rangle$ of the Hamiltonian $\mathcal{H}$ in terms of the actual basis,

(2) replacing $\{|\tilde{\nu}\rangle\}$ with the most important (i.e., largest eigenvalues $w_{\tilde{\nu}}$) eigenstates of the density matrix $\boldsymbol{\rho}$, calculated with respect to $|\psi\rangle$ and $\{|\mu_0\rangle\}$,

(3) changing the additional states $\{|\nu\rangle\}$ in the set $\{|\mu_0\rangle\}$,

(4) orthonormalizing the set $\{|\mu_0\rangle\}$, and returning to step (1).

A simple way to proceed in step (3) is to sweep the bare states $\{|\nu\rangle\}$ through a sufficiently large part of the infinite-dimensional phonon Hilbert space. One can think of the algorithm as ''feeding'' the optimized states with bare phonons, thus allowing the optimized states to become increasingly perfect linear combinations of bare phonon states. Of course the whole procedure converges only for eigenstates of $\mathcal{H}$ at the lower edge of the spectrum, since usually the spectrum of a Hamiltonian involving phonons has no upper bound. The applicability of the algorithm was demonstrated in Ref. 5 with the Holstein model [i.e., $U=0$ in Eq. (1)] as an example.

When we implemented the above algorithm together with a Lanczos exact diagonalization method for our systems of interest, we found two objections against the above choice of an optimized basis: (i) the basis is not symmetric under the symmetry operations of the Hamiltonian (e.g., translations), and (ii) the phonon Hilbert space is still large ($D_{\rm ph}=Mm^{N-1}$, where M is the dimension at the large site), since we usually need more than one optimized state per site.

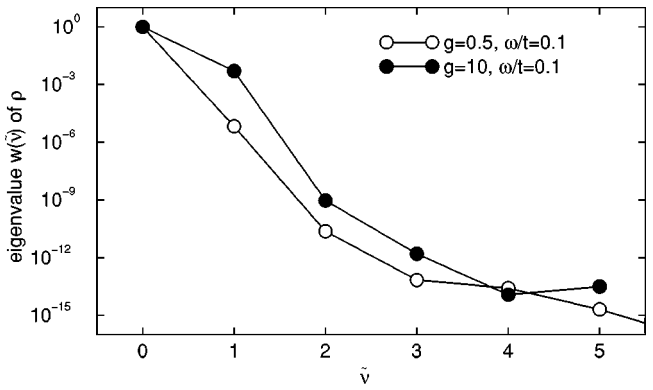

FIG. 1. Eigenvalues $w_{\tilde{\nu}}$ of $\boldsymbol{\rho}$ calculated with the ground state of the Holstein model of spinless fermions for weak and strong coupling. For larger $\tilde{\nu}$ the eigenvalues are close to the numerical precision ($\approx 10^{-14}$), explaining the flattening.

The first problem is solved by including all those states into the phonon basis that can be created by symmetry operations, and by calculating the density matrix in a symmetric way, i.e., by adding the density matrices generated with respect to every site, not just site $i=0$. Concerning the second problem we note that the eigenvalues $w_{\tilde{\nu}}$ of the density-matrix $\boldsymbol{\rho}$ decrease approximately exponentially, see Fig. 1. If we interpret $w_{\tilde{\nu}}\sim\exp(-a\tilde{\nu})$ as the probability of the system to occupy the corresponding optimized state $|\tilde{\nu}\rangle$, we immediately find that the probability for the complete phonon basis state $\otimes_{i=0}^{N-1}|\tilde{\nu}_i\rangle$ is proportional to $\exp(-a\Sigma_{i=0}^{N-1}\tilde{\nu}_i)$. This is reminiscent of the energy cut-off discussed above, and we therefore propose the following choice of phonon basis states at each site,

$$\forall\ \ i:\quad \{|\mu_i\rangle\}=\mathrm{ON}(\{|\mu\rangle\}) \tag{6}$$

$$|\mu\rangle=\begin{cases}\text{opt. state}\quad |\tilde{\nu}\rangle, & 0\leq\mu<m\\ \text{bare state}\quad |\nu\rangle, & m\leq\mu<M\end{cases} \tag{7}$$

and for the complete phonon basis $\{\otimes_{\Sigma_i\mu_i<M}|\mu_i\rangle\}$, yielding $D_{\rm ph}=\binom{N+M-1}{N}$. Implementation of this optimization procedure together with our existing Lanczos diagonalization code[6] allows the study of interacting electron/spin-phonon systems in a much larger parameter space without reaching the limits of available supercomputers.

To demonstrate the power of the method, in the following we address two frequently discussed problems: bipolaron formation and Luttinger liquid behavior in 1D polaronic metals.

Bipolarons in the 1D Holstein-Hubbard model. Bipolaron formation has been the subject of numerous studies over the last decades, stimulated for instance by the discovery of high-T_c cuprates, and the belief that the interplay between strong electron-phonon and electron-electron interactions plays a significant role in these highly correlated materials.[7] Nevertheless the influence of the Hubbard interaction U on bipolaron formation is still not completely understood. Beside bipolaron formation itself, an interesting open question is the transition between two bipolaronic regimes, namely the intersite and the onsite bipolaron. Since the Hubbard interaction U and the electron-phonon interaction compete, we

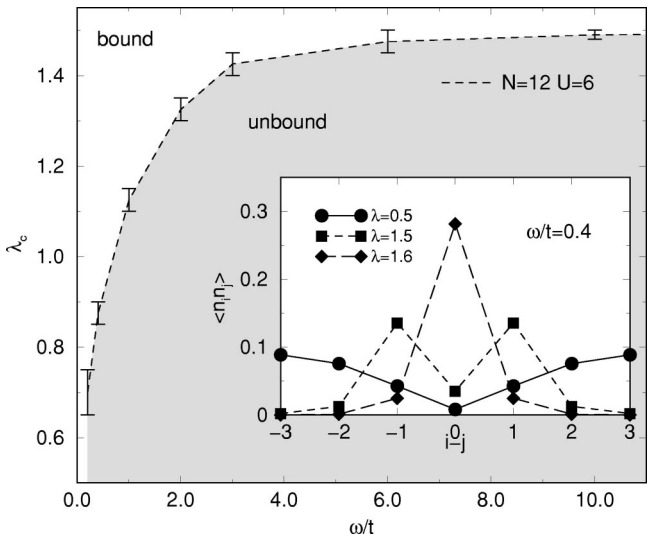

FIG. 2. Critical coupling for bipolaron formation at fixed $U/t=6.0$ and system size $N=12$; inset: $\langle n_i n_j \rangle$ correlation in the unbound, intersite and on-site cases for $\omega/t=0.4$, $N=6$.

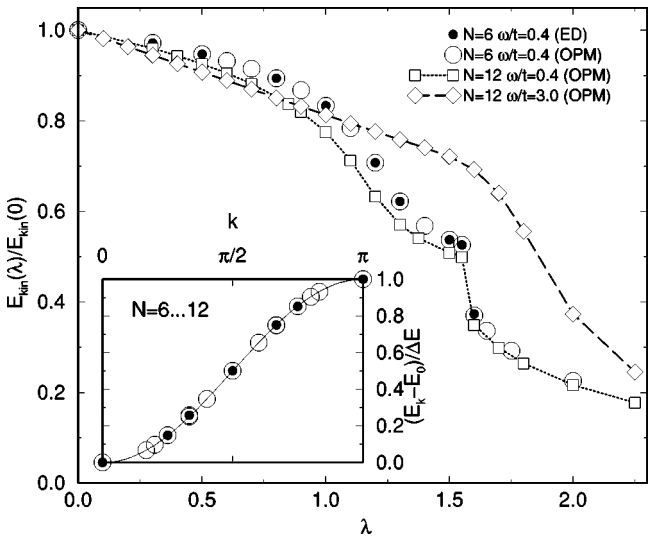

FIG. 3. Bipolaron kinetic energy as a function of coupling strength λ for frequencies $\omega/t=0.4$ and 3.0, comparing optimized (OPM) and bare (ED) phonons. Inset: rescaled bipolaron dispersion at $\omega/t=0.4$, $\lambda=U/(4t)$, where $\Delta E/t=0.0103$ is the bandwidth.

usually need to consider intermediate to strong electron-phonon coupling g, or $\lambda := g^2\omega/(2t)$, making the problem a good testing ground for our optimized phonon algorithm.

In a recent work Bonča *et al.*[8] studied mobile bipolarons in the Hubbard model by means of a variational technique. Their focus is mainly on the U dependence of the transition from unbound polarons to intersite bipolarons and from intersite to onsite bipolarons at intermediate frequencies. These transitions also show a significant ω dependence. Here the adiabatic frequency range is of special interest, since there are no appropriate analytic methods for small but finite frequencies.

Using the optimized phonon approach on lattice sizes up to $N=12$, we calculated the phase diagram for the transition from unbound polarons to bipolarons at fixed U. The critical coupling λ_c was determined by the condition $\Delta=0$, where Δ is the energy difference between the two-particle ground state and twice the one-particle ground state, i.e., $\Delta=E_b-2E_p$. As indicated in Fig. 2, the critical interaction λ_c increases with frequency, reaching $U/(4t)$ in the antiadiabatic strong-coupling limit, where the phonon interaction is instantaneous and the Holstein-Hubbard model maps onto an effective Hubbard model with $U_{\rm eff}=U-4t\lambda$. As $U_{\rm eff}$ becomes negative an onsite bipolaron bound state is formed. In the physically more relevant adiabatic regime, however, the retardation effect of the electron-phonon interaction may favor the formation of more extended electron bound states:[9] due to the time delay the second electron can take advantage of the lattice distortion left by the first one, still avoiding direct Coulomb repulsion. Increasing the electron-phonon coupling the density-density correlation signals the transition to such an intersite bipolaron followed by the formation of an onsite bipolaron (see inset). Most notably, the behavior of the kinetic energy indicates that the intersite bipolaron is a *mobile* quasiparticle even though its effective mass is substantially enhanced (cf., Fig. 3). This is reminiscent of the behavior of large bipolarons in models with long-range (Fröhlich-type) electron-lattice interaction. Another striking property is the almost perfect cosine band dispersion of the nearest-neighbor intersite bipolaron (see inset). This free-particle, like behavior, is indicative of a vanishing residual bipolaron-phonon interaction and can be attributed to the fact that the onsite and nearest-neighbor electron-phonon correlations are about the same size.[10] In contrast the onsite bipolaron is almost immobile (for $\lambda=1.6$ and $\omega/t=0.4$ the renormalized bandwidth becomes smaller than 10^{-3}; cf., also the sharp drop in $E_{\rm kin}$).

Luttinger liquid behavior. The Holstein model of spinless fermions is defined by omitting the electron spin σ and consequently the Coulomb interaction U in Hamiltonian (1). In one dimension and at half filling, depending on the coupling strength g, this model undergoes a transition from a gapless metallic phase to a Peierls distorted phase with a gap between the ground-state and lowest excitations. Details of this transition and the properties of the different phases were studied with several methods over the last years.[11–14] One interesting aspect is the description of the metallic phase in terms of an effective Luttinger model, which, according to the ''Luttinger liquid hypothesis'' of Haldane,[15] should be an universal picture for the low-temperature properties of all one-dimensional metals. The two parameters of the Luttinger model, the renormalized Fermi velocity u_ρ and effective coupling constant K_ρ, can be determined through the scaling behavior of the ground-state energy E_0 and the energy of charge excitations $E_{\pm 1}$ with respect to the system size N:

$$\frac{E_0(N)}{N}=\epsilon_\infty-\frac{\pi u_\rho}{6N^2},\quad E_{\pm 1}(N)-E_0(N)=\frac{\pi u_\rho}{2K_\rho N}. \tag{8}$$

In a recent work[14] we used a variational method to calculate eigenstates and the resulting Luttinger parameters for the Holstein model at half filling. Unfortunately the method failed to give consistent results especially for K_ρ in the antiadiabatic regime of large frequencies $\omega \gg t$ where the Holstein model can be well described by second order perturbation theory, leading to an effective XXZ spin model[11] with known Luttinger parameters. For large frequencies the XXZ

model, as well as Monte Carlo[12] and density-matrix renormalization group (DMRG)[13] calculations for the Holstein model, yield $K_\rho < 1$ corresponding to a repulsive interaction. If K_ρ, starting with the value 1 for the noninteracting case, reaches $\frac{1}{2}$ with increasing coupling strength, the model undergoes a Kosterlitz-Thouless transition to a gapped phase. In contrast, the variational technique yields an attractive interaction ($K_\rho > 1$) for all frequencies. Lanczos diagonalization without optimized phonons could not resolve this unsatisfying situation, because the Hilbert space required to calculate the requested eigenstates with sufficient precision is far too large for all available computers.

Even with the above choice of an optimized phonon basis we need many states ($M \gtrsim N$), since at large electron-phonon coupling we have to cover two different distortion patterns for an empty or occupied site. We therefore implemented a more sophisticated variant of the above algorithm, using two different sets of optimized phonon states, one for each possible fermion occupation number (cf., Ref. 5). Together with the cut-off, this results in a further reduction of the Hilbert space ($M \sim N/2$), which is required for the diagonalization of larger systems. It is worth noting that this advantage is gained at the expense of a less sparse matrix, since every hopping is connected with the projection of the actual phonon state onto the other basis set. Hence, for more complicated models, such as Jahn-Teller problems with two or three phonon modes per site, the diagonalization can be time consuming.

In Fig. 4 we show the Luttinger parameters we found by scaling the energies for system sizes up to $N = 10$. In the antiadiabatic frequency range the renormalized Fermi velocity u_ρ is drastically supressed within the metallic phase, while for low phonon frequencies it remains almost unchanged up to the phase transition. A very interesting result

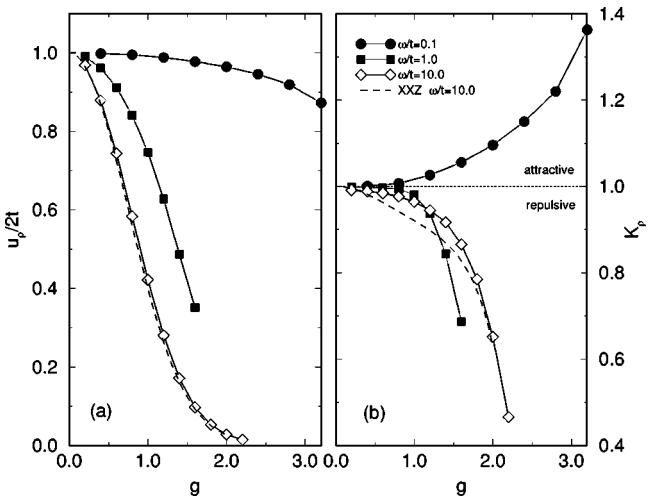

FIG. 4. Luttinger liquid parameters for the Holstein model of spinless fermions at phonon frequencies $\omega = 0.1$, 1.0, and 10.

is the changing character of the interaction below $\omega \sim t$. For small frequencies the effective fermion-fermion interaction is attractive, while it is repulsive for large frequencies, i.e., there is a transition point, depending on g and ω, where the model is free in lowest order.

In conclusion, we have proposed an advanced phonon optimization algorithm for application in Lanczos diagonalization, and demonstrated its reliability for two strongly interacting electron-phonon systems.

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