Contrasting unitary transformations for the standard bipolaron model

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The well known bipolaron prototype model [two sites, two electrons, two spins, single oscillator (''222 model'')] is considered, involving a Fröhlich-type electron-phonon interaction and a Hubbard term. As in the one-electron case a Fulton-Gouterman transcription leads to a symmetry-adequate definition of the oscillatory companion functions of the two-electron base vectors. Trial expressions for these oscillatory functions are generated by means of three unitary operators (''displacive, squeezing, and reflective'') acting within the vibrational subspace. Several different combinations of these operators are used as generators. Comparing the optimized results with the results of an exact numerical diagonalization, it is found that one of these combinations, involving all three basic operators is the superior one in all coupling regions. Specifically it is found that in the intermediate and strong coupling regimes the ''reflection'' operation is essential. A combination of squeezing and displacement alone, as frequently used in literature, cannot reproduce the two-peaked nature of the vibrational wave functions. [S0163-1829(99)01129-7]

I. INTRODUCTION

The investigations on the famous Landau-polaron problem[1] have by now a history of more then six decades. In this long time period the intensity of investigations of this problem had several peaks, and recently the problem has become virulent again in the context of local pairing concepts, e.g., in high-T_c materials.[2–4] In these concepts renewed attention is given to the Schafroth model of superconductivity,[5] which preceded the BCS theory, and to the work of Alexandrov and Ranninger.[6] Since the high-T_c materials are of highly dielectric nature, it seems suggestive to consider a degenerate gas of polarons or bipolarons, which has been done by Mott.[7] A very recommendable introduction to the physical background for a polaron theory of high-temperature superconductors is presented in an article of Mott.[8]

Already in early stages of the polaron discussion unitary transformations have been employed. In particular the very fruitful papers of Sander and Shore[9] and of Lang and Firsov[10] are noteworthy. In the paper of Sander and Shore the two-site one-electron problem has been considered by means of the Fulton-Gouterman (FG) transformation.[11,12] In this way the problem is reduced to one of the oscillatory subspace. The great virtue of this reduction is the direct insight into the dominant antagonistic tendencies of the coupled electron-phonon dynamics. The latter has been analyzed in more detail in our group.[13] The main outcome of the work of Shore and Sander was an important result about the structural form of the vibrational wave function of the ground state. It turned out that the form consisted of a displaced dominant peak and a smaller mirror image of this peak. It was this structural form which made it possible to analyze the transition from a non-self-trapped to a self-trapped character of the electron. In particular the outcome was that the transition is not of a jump character but displays a smooth nature. An approach also somewhat in the spirit of Sander and Shore has recently been presented by La Magna and Pucci[14] for an extended Holstein-chain and Fehske *et al.*[15] have investigated the spectral properties of the two-dimensional Holstein polaron.

As regards the two-electron problem the state of art is less advanced. As a prototype model for the bipolaron problem very often the two-site two-electron two-spin (''222'') model has been handled. Worth mentioning is also a three-site two-mode model, involving both an infrared- and a Raman-active mode. This type of model has been treated by Bishop and Salkola.[16] In the present work we confine ourselves to the ''222 model.'' [17] This model is accessible to exact numerical treatment and therefore may be exploited to evaluate the power of approximative methods. In the present work we specifically analyze the utilization of unitary transformations for the approximate solution of the problem.

In Sec. II the 222 model is introduced und subjected to the Fulton-Gouterman transformation (FGT). This establishes the exact equations for the respective oscillatory companion functions for the electronic base vectors. In Sec. III the exact numerical form of these companion functions and of their characteristic properties is presented. In Sec. IV the oscillatory functions are generated by means of unitary operators and optimized. By comparing the results with the exact ones this allows for an assessment of the utility of unitary operators, which is discussed in Sec. V.

II. THE TWO-SITE BIPOLARON MODEL

We considered a prototype model which frequently has been discussed in literature (i.e., Refs. 18–25). The Hamiltonian is given by

$$H_d = H_{\mathrm{ph}} + H_e + H_{e\text{-}e} + H_{e\text{-ph}}, \tag{1}$$

$$H_{\mathrm{ph}} = \frac{\hbar\Omega}{2} \cdot (P^2 + Q^2), \tag{2}$$

$$H_e = -T \cdot \hbar\Omega \cdot (\sigma_{x\uparrow} + \sigma_{x\downarrow}), \tag{3}$$

$$H_{e\text{-}e}=U\cdot\hbar\Omega\cdot\left(\frac{1}{2}\hat{n}_{\uparrow}\hat{n}_{\downarrow}+2\sigma_{z\uparrow}\sigma_{z\downarrow}\right), \tag{4}$$

$$H_{e\text{-ph}}=\hbar\Omega\cdot D\cdot Q\cdot(\sigma_{z\uparrow}+\sigma_{z\downarrow}). \tag{5}$$

The first term represents an effective harmonic oscillator and the second the electronic transfer. The third and fourth terms stand for the Coulomb-Hubbard and the electron-oscillator interaction, respectively.

In this model it is assumed that there are two equivalent atomic sites, each of which has one electronic orbital of the same nature. The system displays inversion symmetry and there is an effective oscillatory coordinate Q (momentum P) of odd parity nature. The electronic creators (annihilators) are designated by c_{lj}^{+}, c_{rj}^{+} (c_{lj}, c_{rj}), j standing for the two spin directions $(j=\uparrow,\downarrow)$ and l, r insinuating ''left'' and ''right.'' We further have introduced the abbreviations

$$n_{lj}=c_{lj}^{+}c_{lj}, \quad n_{rj}=c_{rj}^{+}c_{rj}, \quad n_j=n_{lj}+n_{rj}, \tag{6}$$

$$\sigma_{xj}=\frac{1}{2}(c_{lj}^{+}c_{rj}+c_{rj}^{+}c_{lj}), \quad \sigma_{yj}=\frac{1}{2i}(c_{lj}^{+}c_{rj}-c_{rj}^{+}c_{lj}),$$

$$\sigma_{zj}=\frac{1}{2}(c_{lj}^{+}c_{lj}-c_{rj}^{+}c_{rj}). \tag{7}$$

The quantities (7) display spinlike commutation relations. We further introduce reflection operators given by

$$R=R_{\mathrm{el}}R_Q=R_QR_{\mathrm{el}},$$

with

$$(R_{\mathrm{el}}R_Q)^{+}=R_{\mathrm{el}}R_Q, \quad (R_{\mathrm{el}}R_Q)^2=1. \tag{8}$$

R_{el} and R_Q representing respectively reflections in the electronic and the phononic subspaces. They have the basic properties

$$R_QQ=-QR_Q,$$

$$R_QP=-PR_Q,$$

with

$$R_Q^{+}=R_Q, \quad R_Q^2=1 \tag{9}$$

$$R_{\mathrm{el}}c_{lj}=c_{rj}R_{\mathrm{el}}, \quad R_{\mathrm{el}}c_{rj}=c_{lj}R_{\mathrm{el}},$$

$$R_{\mathrm{el}}c_{lj}^{+}=c_{rj}^{+}R_{\mathrm{el}}, \quad R_{\mathrm{el}}c_{rj}^{+}=c_{lj}^{+}R_{\mathrm{el}},$$

with

$$R_{\mathrm{el}}^{+}=R_{\mathrm{el}}, \quad R_{\mathrm{el}}^2=1 \tag{10}$$

and

$$[R_{\mathrm{el}}R_Q,H]_{-}=0. \tag{11}$$

In the model it is further assumed that the number of the electrons is fixed to $N=2$. From the considered four one-electron states we deduce a two-electron basis given by six functions

$$\left.\begin{aligned}|l\rangle&=c_{l\uparrow}^{+}c_{l\downarrow}^{+}|\text{vacuum}\rangle\\ |r\rangle&=c_{r\uparrow}^{+}c_{r\downarrow}^{+}|\text{vacuum}\rangle\\ |h\rangle&=\frac{1}{\sqrt{2}}(c_{l\uparrow}^{+}c_{r\downarrow}^{+}-c_{l\downarrow}^{+}c_{r\uparrow}^{+})|\text{vacuum}\rangle\end{aligned}\right\}\ \text{singlet-states}\ (S=0, \quad S_z=0) \tag{12}$$

$$\left.\begin{aligned}|t\rangle&=\frac{1}{\sqrt{2}}(c_{l\uparrow}^{+}c_{r\downarrow}^{+}+c_{l\downarrow}^{+}c_{r\uparrow}^{+})|\text{vacuum}\rangle\\ |t,+1\rangle&=c_{l\uparrow}^{+}c_{r\uparrow}^{+}|\text{vacuum}\rangle\\ |t,-1\rangle&=c_{l\downarrow}^{+}c_{r\downarrow}^{+}|\text{vacuum}\rangle\end{aligned}\right\}\ \text{triplet-states}\ (S=1, \quad S_z=0,\pm1). \tag{13}$$

Since the Hamiltonian involves no spin-flip terms we may handle the solutions in the triplet and the singlet subspaces separately. Specifically, we will restrict our results to the solutions in the singlet subspace, since in the usual parameter constellations the lowest states are in this subspace. The exact total eigenfunctions then must be of the Wigner form:[26]

$$|\Psi_g\rangle=\left[\frac{1}{\sqrt{2}}\Big(|l\rangle\,\phi_l(Q)+|r\rangle\cdot\underbrace{R_Q\phi_l(Q)}_{=\phi_r(Q)}\Big)+|h\rangle\,\phi_h(Q)\right] \tag{14}$$

with

$$\langle\phi_l(Q)|\phi_l(Q)\rangle+\langle\phi_h(Q)|\phi_h(Q)\rangle=1,$$

$$R_Q\phi_h(Q)=\phi_h(Q) \tag{15}$$

and

$$|\Psi_u\rangle=\frac{1}{\sqrt{2}}(|l\rangle-R_Q|r\rangle)\,\phi_u(Q), \tag{16}$$

with

$$\langle\phi_u(Q)|\phi_u(Q)\rangle=1, \tag{17}$$

i.e., of a form displaying a parity $p=g$, u, such that

$$R|\Psi_p\rangle=p|\Psi_p\rangle. \tag{18}$$

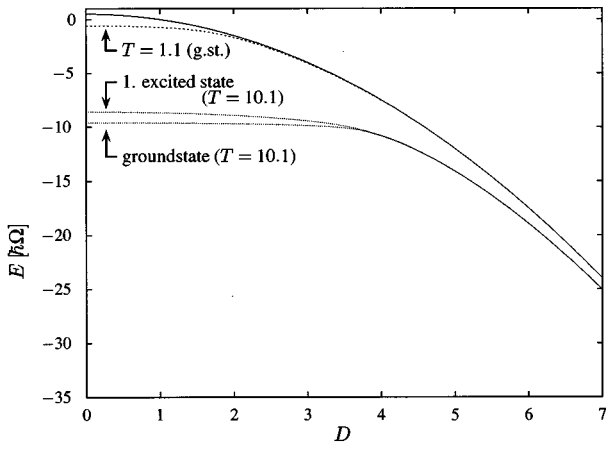


FIG. 1. Bipolaron model without Hubbard term (''Holstein model,'' $U=0$). Matrix diagonalization: ground state E as a function of the electron-phonon coupling strength D for the transfer values $T=1.1$(— — —) and $T=10.1$(· · · ·). For $T=10.1$ there is a tendency to degeneration witht he first excited state for higher D values. The limiting case $T=0.0$(——) is also drawn.

The vibrational functions $\phi_l(Q)$ and $\phi_h(Q)$ satisfy coupled eigenvalue equations (''Fulton-Gouterman equations''), which can be found via a variational principle[11,27] or by a unitary transformation,[28,12] and read

$$[H_{\rm ph}+\hbar\Omega\cdot U+\hbar\Omega\cdot DQ]\phi_l(Q)-\hbar\Omega\cdot T\cdot\phi_h(Q) = E_g(Q)\phi_l(Q), \tag{19}$$

$$H_{\rm ph}\phi_h(Q)-\hbar\Omega\cdot\frac{T}{2}\cdot(1+R_Q)\phi_l(Q)=E_g(Q)\phi_h(Q), \tag{20}$$

$$[H_{\rm ph}+\hbar\Omega\cdot U+\hbar\Omega\cdot DQ]\phi_u(Q)=E_u(Q)\phi_u(Q). \tag{21}$$

This transcription also is known as the Fulton-Gouterman transformation.[11,28,12] We note that the two vibrational functions $\phi_l(Q)$ and $\phi_h(Q)$ pertaining to the even parity case have to be calculated by solving a coupled system of eigenvalue equations (19) and (20), and we emphasize that the transcriptions (19)–(21) represent an exact substitute of the original Schrödinger equation.

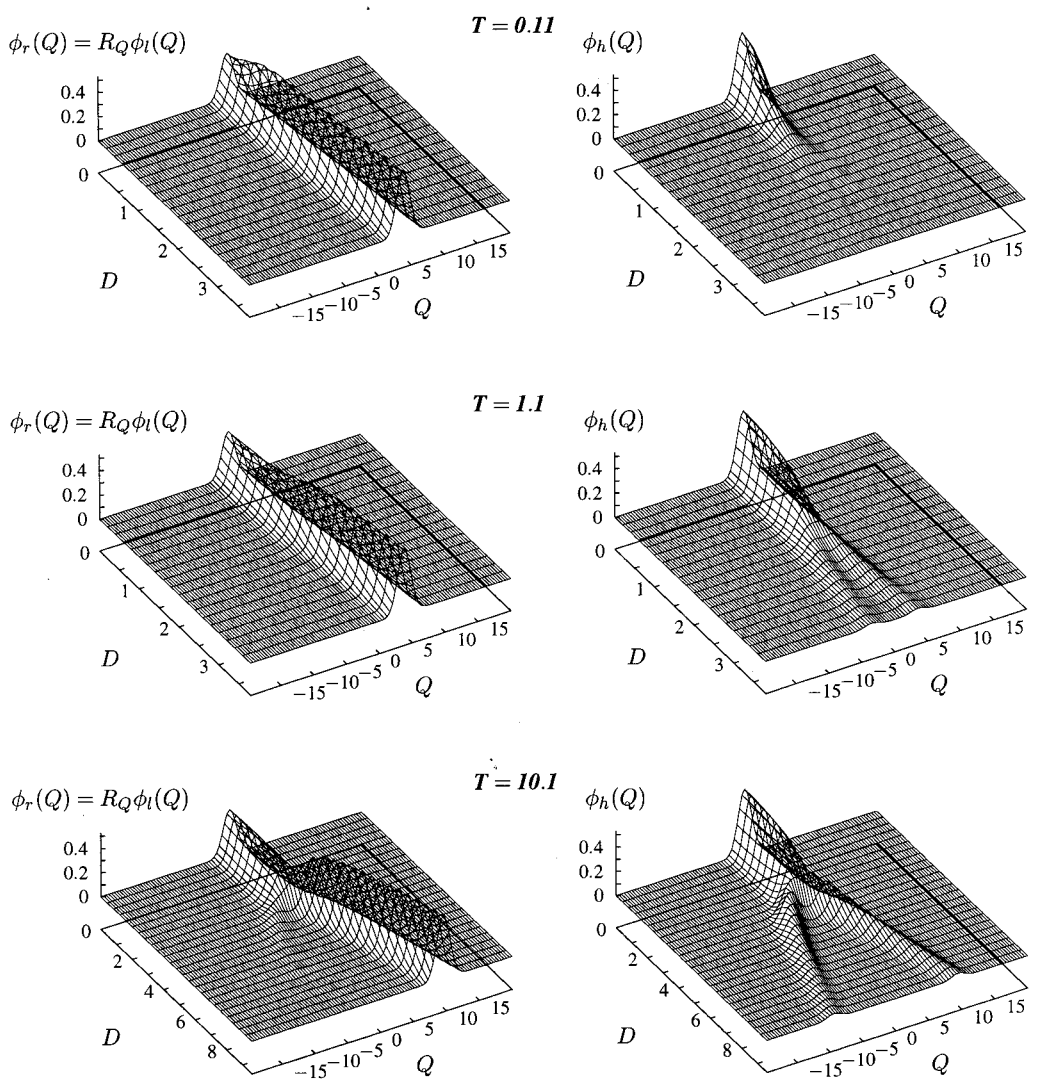


FIG. 2. Biopolaron model without Hubbard term (''Holstein model,'' $U=0$). Exact numerical diagonalization: phononic ground state wave function $\phi_r(Q)=R_Q\phi_l(Q)$ and $\phi_h(Q)$ for $T_0=0.11, T_0=1.1$, and $T_0=10.1$ in a Q-D display.

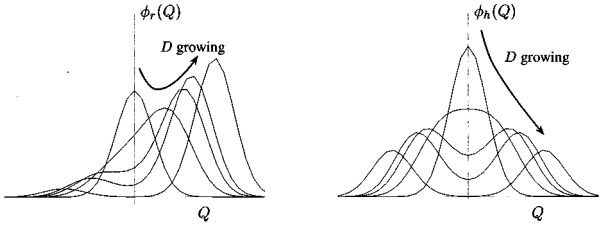


FIG. 3. Bipolaron model without Hubbard term (''Holstein model,'' $U=0$). Matrix diagonalizaiton: characteristics of the phononic ground state wave function $\phi_r(Q)=R_Q\phi_l(Q)$ and $\phi_h(Q)$ for different values of the electron-phonon coupling parameter D and the transfer parameter $T=10.1$.

In the limiting cases for the model a number of analytical results are known from literature. We refrain from writing down these properties but refer to the literature.[22]

III. EXACT NUMERICAL SOLUTION

In this work we intend to analyze contrasting combinations of unitary transformations with regard to their ability of yielding a good diagonalization of the Hamiltonian, i.e., of generating good trial wave functions. To quantitatively estimate this ability we make reference to the numerically exact ground state properties.

Since the combined electron-oscillator problem of the singlet subspace contains only three electronic base functions ($|l\rangle,|r\rangle,|h\rangle$) and a single oscillator which may be represented by the eigenbase $\{\varphi_n(Q)\}$ of $H_{\rm ph}=(\hbar\Omega/2)(P^2+Q^2)$, the considered Hamiltonian may be handled exactly by numerical diagonalization. For the considered parameters a base of the lowest 250 functions $\varphi_n(Q)$ provides a sufficient accuracy. The groundstate of the 222 model also has been treated in Ref. 24 in a numerically exact manner. In a more recent work Alexandrov *et al.*[29] have employed a full numerical diagonalisation of the 222 model to calculate dynamical properties (optical conductivity). In our study we will exploit only the ground state properties. These are documented in the following figures. A test of the analyzed unitary transformations with regard to dynamical properties is in progress.

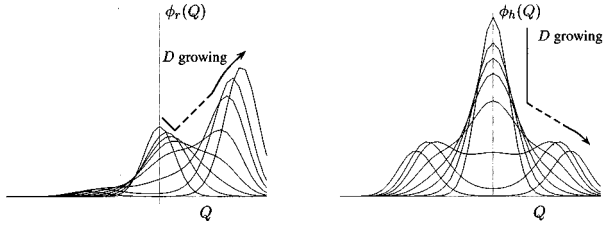


FIG. 5. Hubbard bipolaron model ($U=12.6$). Matrix diagonalization: characteristics of the phononic ground state wave function $\phi_r(Q)=R_Q\phi_l(Q)$ and $\phi_h(Q)$ for different values of the electron-phonon coupling parameter D and the transfer parameter $T=10.1$.

A. Holstein model ($U=0$)

Figure 1 shows the behavior of the ground state, if the Hubbard term is disregarded ($U=0$, ''Holstein model''). It illustrates the ''polaronic'' effects, i.e., the capture of a phononic cloud by the electron with increasing coupling, causing a lowering of the energy. The corresponding oscillatory wave functions, as defined in Eq. (14), are illustrated in Fig. 2. Both vibrational companions $\phi_r(Q)=R_Q\phi_l(Q)$ and $\phi_h(Q)$ of the electronic wave functions respectively display three outstanding characteristics.

1. Characteristics of the function $\phi_r(Q)=R_Q\phi_l(Q)$ in the Holstein model

From Fig. 3 we observe the following characteristics.

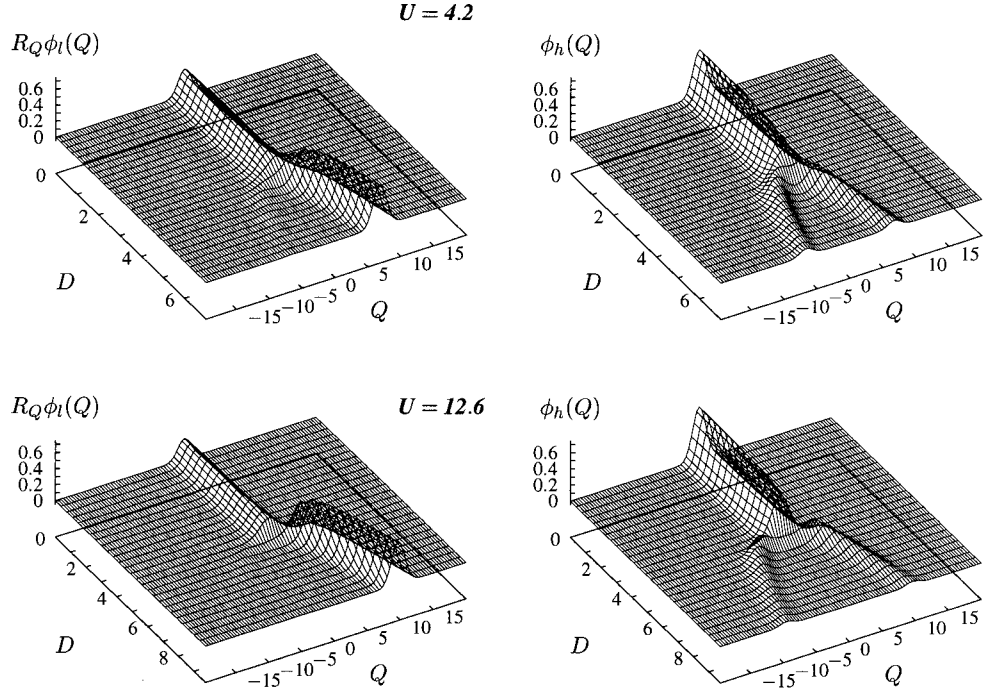


FIG. 4. Hubbard bipolaron model ($U\neq 0$). Matrix diagonalization: phononic ground state wave function $\phi_r(Q)=R_Q\phi_l(Q)$ and $\phi_h(Q)$ for $U=4.2$ and $U=12.6$ with $T=10.1$ depending on the electron-phonon coupling D.

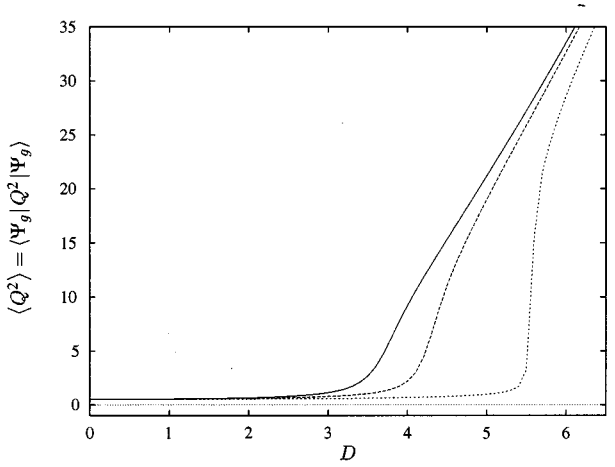


FIG. 6. Bipolaron model Matrix diagonalization. Expectation value of the squared displacement Q for the ground state wave functions $|\Psi_g\rangle$ for $T=10.1$ and $U=0.0$(——),$U=4.2$(- - -), and $U=12.6$(— — —). The sharpening of the transition to ''localization'' with growing U may be noted.

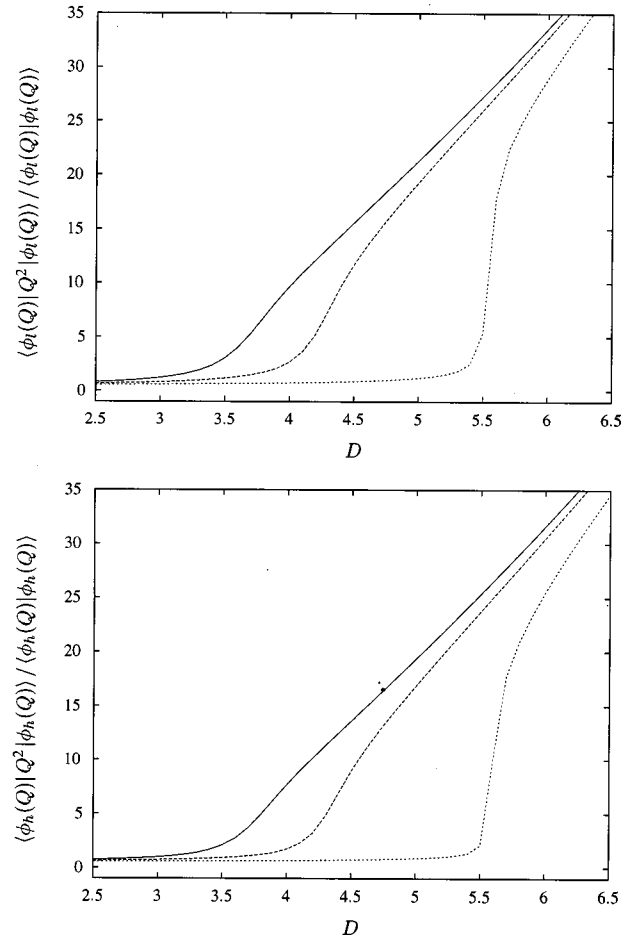


FIG. 7. Bipolaron model. Matrix diagonalization. Squared displacement Q for the oscillatory ground state companion wave functions [see Eq. (14)]: $\phi_l(Q)$ (upper figure) and $\phi_h(Q)$ (lower figure) (ground state) for $T=10.1$ and $U=0.0$(——),$U=4.2$(- - -), and $U=12.6$(— — —).

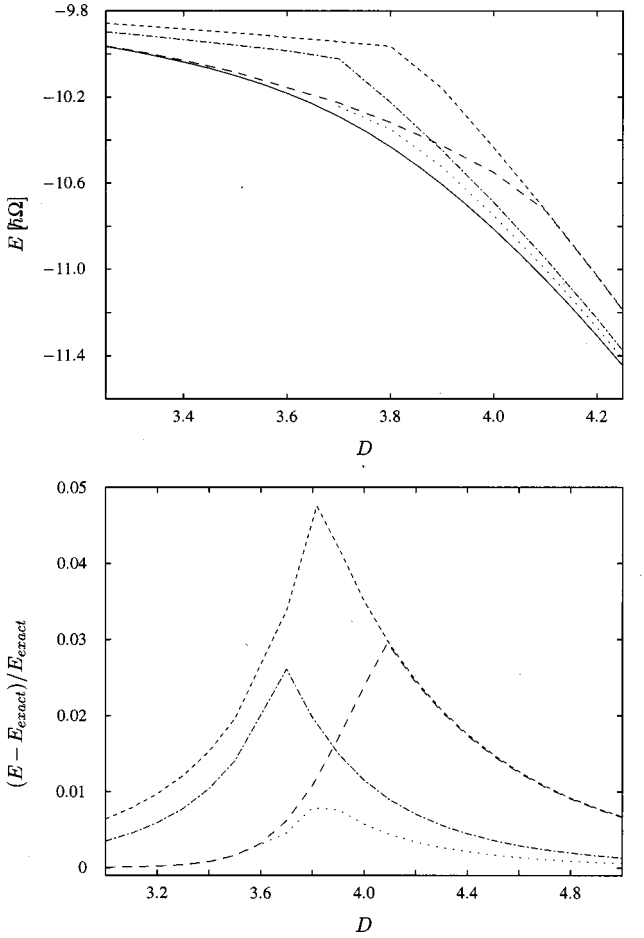


FIG. 8. Holstein-bipolaron model (U=0). Ground state energy (upper figure) and relative deviation from the exact value (lower figure) in dependency of the electron-phonon coupling D for $T=10.1$: displacive ansatz (- - -), squeezing-displacive ansatz (— — —), nonunitary reflective ansatz (— · — · —), and exact value (——).

Displacement (''localization''). As seen from the figure the maximum of $\phi_r(Q)$ shifts to the right with growing electron-phonon coupling D. This shift physically amounts to a tendency of the total wave function versus localization.

Antisqueezing (''broadening''). As also seen from the figure the shape of $\phi_r(Q)$ covers a spatial region which is broadening with growing electron-phonon coupling D. In a *rough approximation* this broadening may be viewed as an ''antisqueezing'' of the undisturbed functional form, and trial functions may generated by means of a ''squeezing transformation'' in combination with a displacement transformation. This has been done in literature.[13,30]

Separation in two parts (''shoulder''). Finally, as also noted from Fig. 3, we observe a splitting of the function $\phi_r(Q)$ into a dominant peak on the right side and a minor mirror image peak on the left side. This separation is getting more and more pronounced for growing coupling values D, whereas for smaller coupling values D the dominant peak only displays a ''shoulder'' on its left side.

2. *Characteristics of the function $\phi_h(Q)$ in the Holstein model*

We find the following characteristics (see Fig. 3).

Symmetric structure in Q space [see Eq. (15)].

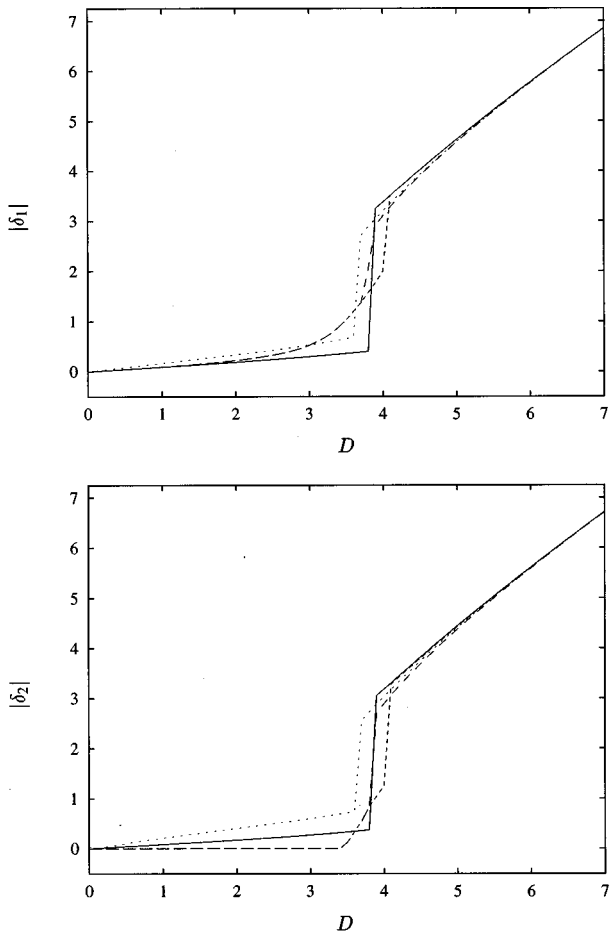


FIG. 9. Holstein-bipolaron model ($U=0$). Displacement parameters δ_1 and δ_2 in dependency of the electron-phonon coupling parameter D for $T_0=10.1$: displacive ansatz (——), squeezing-displacive ansatz (- - -), nonunitary reflective ansatz (— — —), unitary reflective-displacive ansatz ($\cdot\ \cdot\ \cdot\ \cdot$).

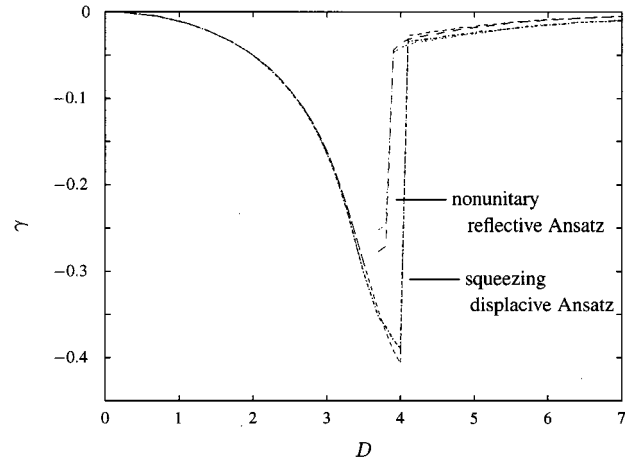


FIG. 10. Holstein-bipolaron model ($U=0$). Squeezing parameters γ_1 and γ_2 in dependency of the electron-phonon coupling parameter D for $T_0=10.1$: squeezing-displacive ansatz γ_1(— — —) and γ_2(—·—·—·—), and nonunitary reflective ansatz γ_1(— — —) and γ_2(—·—·—·—).

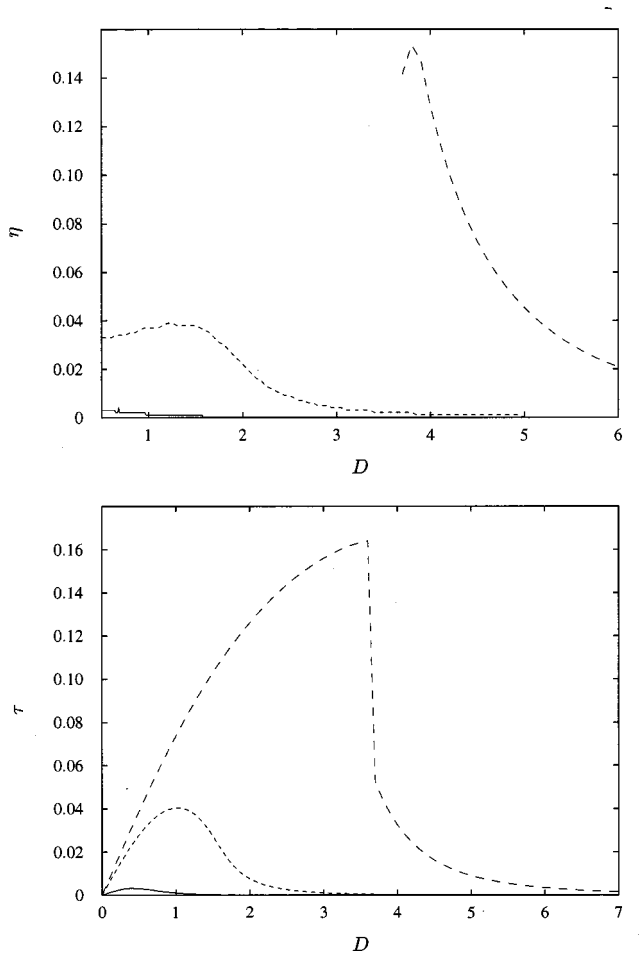


FIG. 11. Holstein-bipolaron model ($U=0$). Parameters η and τ [see Eqs. (43) and (44)] in dependency of the electron-phonon coupling parameter D for $T_0=0.11$(——), $T_0=1.1$(- - -), and $T_0=10.1$(— — —).

Antisqueezing (''broadening''). For growing values of D the regional extension of $\phi_h(Q)$ increases and the remarks referring to $\phi_r(Q)$ apply also here. In a *rough approximation* the broadening again can be generated by means of a ''squeezing transformation.''

Separation in two parts (''two peaks''). From Fig. 3 we observe an indentation in the center and a separation in two peaks displaced in opposite directions. For growing values of D we note the generation of a small minimum, which eventually leads to a separation of $\phi_h(Q)$ into two equal functional forms and amounts to localization.

B. Role of the Hubbard term

Figure 4 shows the oscillatory wave functions $\phi_r(Q)$ and $\phi_h(Q)$ in a three-dimensional display if the Hubbard parameter U is nonzero. The main features of the functions of the ''Holstein'' model are still present. However, there is one interesting effect, which is the sharpening of the transition from the nonlocalized behavior to the localized behavior. This is manifest in the sudden shift of the dominant peak of $\phi_r(Q)$ (see lower left figure) or in the sharp upcome of the two peaks in $\phi_l(Q)$ (see lower right figure), if we compare them with the corresponding figures in the ''Holstein'' model (see Fig. 2).

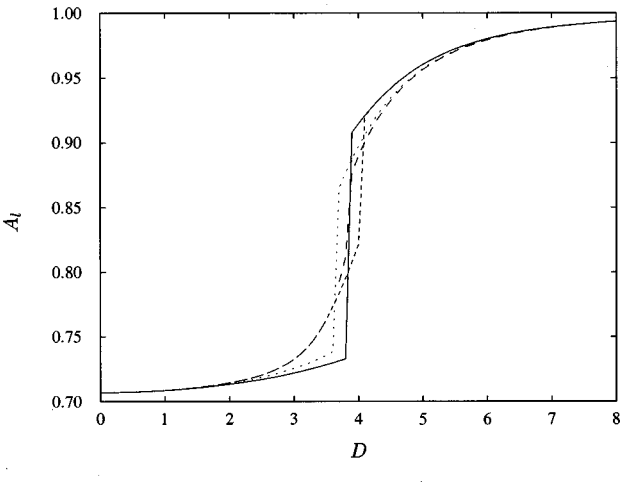


FIG. 12. Holstein-bipolaron model ($U=0$). Parameter A_l [for definition, see Eqs. (27)–(29)] in dependency of the electron-phonon-coupling parameter D for $T_0=10.1$: displacive ansatz (——), squeezing-displacive ansatz (− − −), nonunitary reflective ansatz (- - -), and unitary refective-displacive ansatz (· · · ·).

Figure 5 is the ''Hubbard'' analogue of Fig. 3. Comparing the behavior of $\phi_r(Q)$ in both cases, we notice a qualitative difference. Whereas in the ''Holstein'' cases (Fig. 3) the higher maximum remains on the right hand side, there is a switch in the ''Hubbard'' case, such that the higher peak at small coupling values shows up on the left hand side and then turns to the right hand side. This behavior just illustrates the sharp transition to ''localization'' as described above.

A further illustration of the upcoming sharp transition in the Hubbard case is shown in Fig. 6, where the expectation value of the squared displacement Q for the ground state wave functions is given. In the two subfigures of Fig. 7 the partial expectation value $\langle Q^2\rangle$ with regard to the vibrational functions $\phi_l(Q)$ and $\phi_h(Q)$ are shown.

IV. TRIAL WAVE FUNCTIONS IN THE FULTON-GOUTERMAN PICTURE

A. Variational principle in the Fulton-Gouterman picture

As mentioned in Sec. II the Fulton-Gouterman equations (19)–(21) may be viewed as the manifestation of a variational principle. Using the ansatz (14) the energy expectation value for the even parity solution (ground state) reads

$$E_g=\langle\phi_l|\left[\frac{\hbar\Omega}{2}(P^2+Q^2)+\hbar\Omega\cdot D\cdot Q+\hbar\Omega\cdot U\right]|\phi_l\rangle + \langle\phi_h|\left[\frac{\hbar\Omega}{2}(P^2+Q^2)\right]|\phi_h\rangle - 2\cdot\hbar\Omega\cdot\frac{T}{2}\cdot(A_g+B_g), \tag{22}$$

with

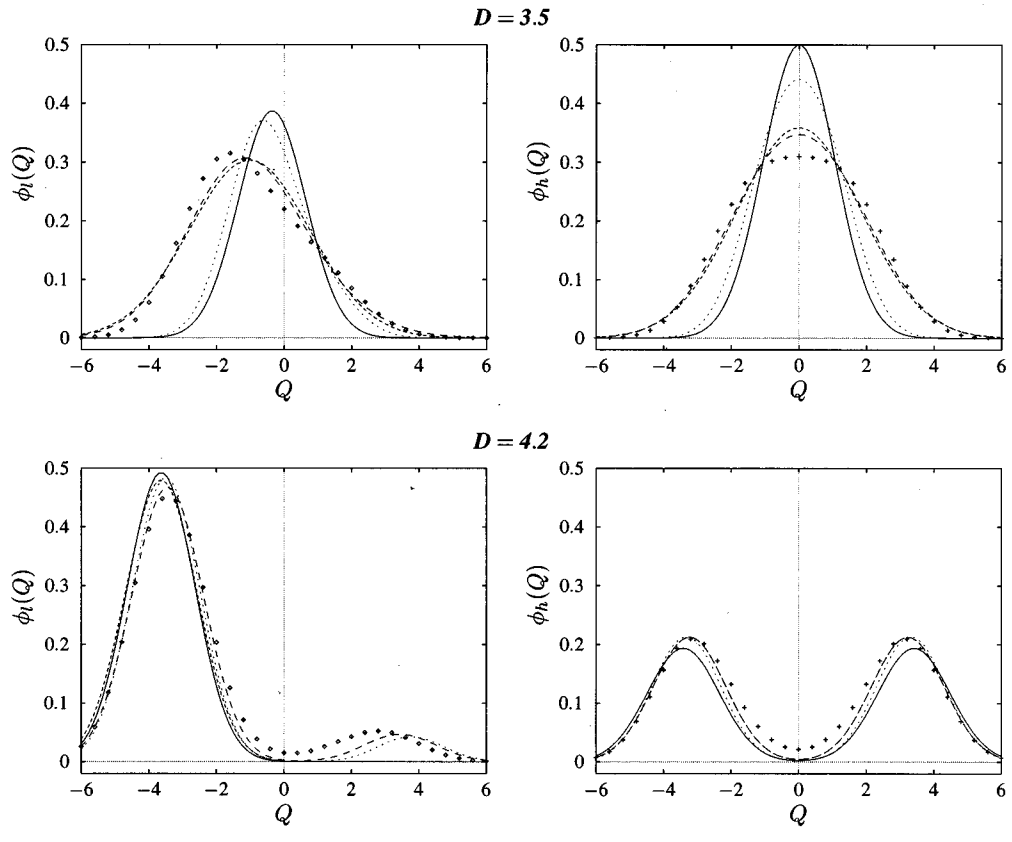


FIG. 13. Holstein-bipolaron model ($U=0$). Phononic partner ground state wave functions $\phi_l(Q)$ and $\phi_h(Q)$[see Eq. (14)] for the electron-phonon coupling parameter $D=3.5$ and $D=4.2$ for $T=10.1$: displacive ansatz (——), squeezing-displacive ansatz (- - -), nonunitary reflective ansatz (− − −), and unitary reflective-displacive ansatz (· · · ·). The exact result from matrix diagonalization is shown by dots (◇ / +).

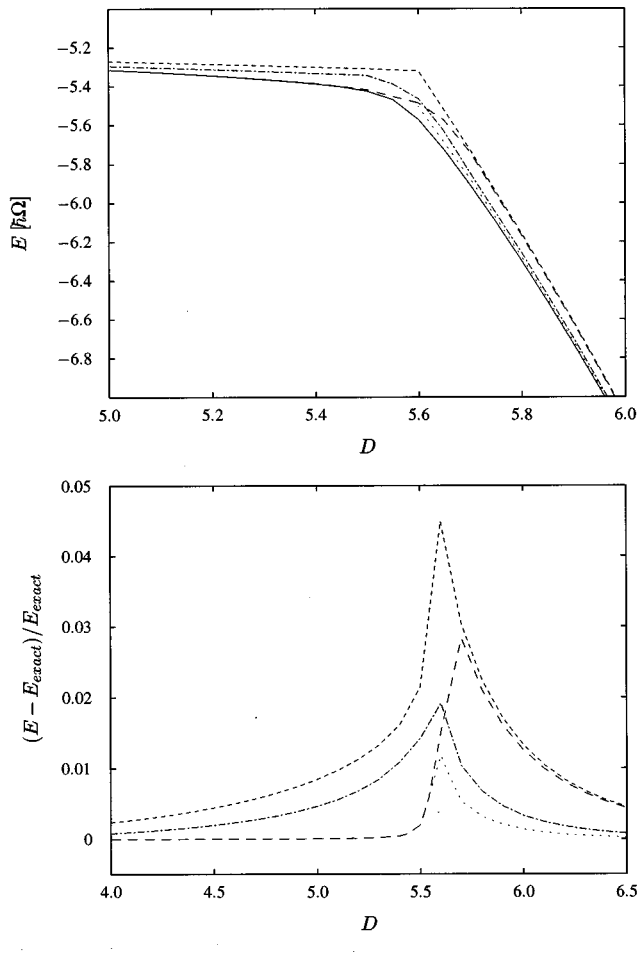


FIG. 14. Hubbard-bipolaron model ($U\neq 0$). Ground state energy (upper figure) and relative deviation from the exact value (lower figure) in dependency of the electron-phonon coupling D for $T=10.1$ and $U=12.6$: displacive ansatz (- - -), squeezing-displacive ansatz (– – –), nonunitary reflective ansatz (· · · ·), unitary reflective-displacive ansatz (—·—·—·—) and exact value (——).

$$A_g=\langle\phi_l|\phi_h\rangle=\langle\phi_h|\phi_l\rangle, \tag{23}$$

$$B_g=\langle\phi_l|R_Q|\phi_h\rangle=\langle\phi_h|R_Q|\phi_l\rangle. \tag{24}$$

Instead of treating the exact eigenvalue equations (19) and (20) we may consider $\phi_l(Q)$ and $\phi_h(Q)$ as trial wave functions and consider the variational principle

$$\delta E_g=\delta E_g(\phi_l(Q),\phi_h(Q))=0, \tag{25}$$

with the conditions

$$\langle\phi_l(Q)|\phi_l(Q)\rangle+\langle\phi_h(Q)|\phi_h(Q)\rangle=1,$$

$$R_Q\phi_h(Q)=\phi_h(Q). \tag{26}$$

The Fulton-Gouterman equation for the odd states $\phi_u(Q)$ simply represent displaced oscillatory equations and may be solved directly.

B. Generation of trial wave functions

The most convenient and elegant way of generating trial wave functions is by the use of unitary operators:[12]

$$\phi_l(Q)=A_l\cdot U_l\varphi_0(Q), \tag{27}$$

$$\phi_h(Q)=A_h\cdot U_h\varphi_0(Q), \tag{28}$$

where $\varphi_0(Q)$ is the ground state of the undisturbed oscillator Hamiltonian [vid. Eq. (2)]. If this is done the normalization condition (15) reads

$$A_l^2+A_h^2=1 \tag{29}$$

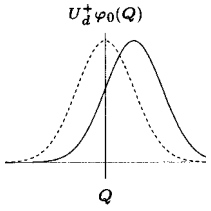
and the variational option (''variational parameters'') is completely incorporated in the choice of the unitary operators U_t and U_e. Most of our trial functions will be chosen in this manner. However we will include also some trial functions not generated in this manner. In particular we will employ also additive combinations of the three unitary transformations.

(1) Displacive transformation (Ref. 12, p. 241): $U_d=e^{i\delta P}$ (δ: real)

$$U_d^+QU_d=Q+\delta, \tag{30}$$

$$U_d^+PU_d=P, \tag{31}$$

$$U_d^+\varphi_0(Q)=e^{-i\delta P}\varphi_0(Q)=\varphi_0(Q-\delta)= \tag{32}$$

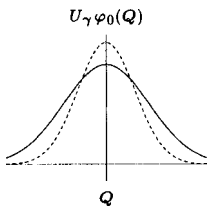


(2) Squeezing transformation (Ref. 12, p. 247): $U_\gamma=e^{i\gamma(PQ+QP)}$ (γ: real)

$$U_\gamma^+QU_\gamma=e^{-2\gamma}Q, \tag{33}$$

$$U_\gamma^+PU_\gamma=e^{2\gamma}P, \tag{34}$$

$$U_\gamma^+\varphi_0(Q)=e^{-i\gamma(PQ+QP)}\varphi_0(Q)=e^{-\gamma}\varphi_0(e^{-2\gamma}Q)=$$



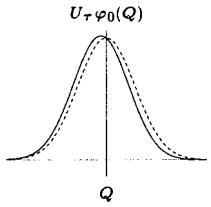
(35)

(3) Reflective transformation (Ref. 31, and Ref. 12, p. 334): $U_\tau = \exp(\tau Q R_Q) = \cos(\tau Q) + \sin(\tau Q) R_Q$ (τ: real)

$$U_\tau^+ Q U_\tau = \cos(2\tau Q)\cdot Q + \sin(2\tau Q)\cdot Q R_Q\,, \tag{36}$$

$$U_\tau^+ P U_\tau = \cos(2\tau Q)\cdot P + \sin(2\tau Q)\cdot P R_Q + i\tau\cdot\sin(2\tau Q) - i\tau\cos(2\tau Q)\cdot R_Q\,, \tag{37}$$

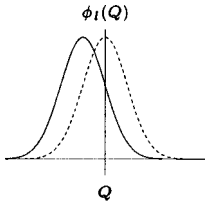
$$U_\tau \varphi_0(Q) = \exp(-\tau Q R_Q)\varphi_0(Q) = \tag{38}$$



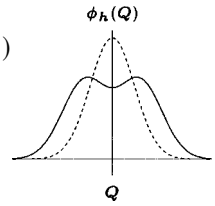
1. Displacive ansatz

The displacive ansatz is as follows:

$$\phi_l(Q) = e^{i\delta_l P}\varphi_0(Q) = \tag{39}$$



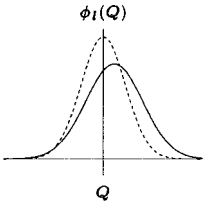
$$\phi_h(Q) = \frac{1}{\sqrt{2(1+\exp(-\delta_h^2))}}(e^{i\delta_h P} + e^{-i\delta_h P})\varphi_0(Q) \tag{40}$$



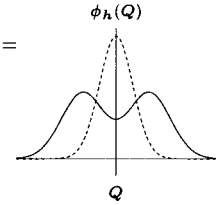
2. Squeezing-displacive ansatz

The squeezing-displacive ansatz is as follows:

$$\phi_l(Q) = e^{i\delta_l P} e^{i\gamma_l(PQ+QP)}\varphi_0(Q) = \tag{41}$$



$$\phi_h(Q) = \frac{1}{\sqrt{2(1+\exp(-\delta_h^2\cdot e^{4\gamma_h}))}}(e^{i\delta_h P} + e^{-i\delta_h P})e^{i\gamma_h(PQ+QP)}\varphi_0(Q) = \tag{42}$$



In passing we mention that the ansatz (41), generated by the product operator $e^{i\delta_l P}e^{i\gamma_l(PQ+QP)}$ yields, after minimization, the same ground state energy as an ansatz respectively generated by the product operator $e^{i\gamma_l(PQ+QP)}e^{i\delta_l P}$ or by the single exponential operator $e^{i\delta_l P + i\gamma_l(PQ+QP)}$. For brevity we refrain from showing this in detail.

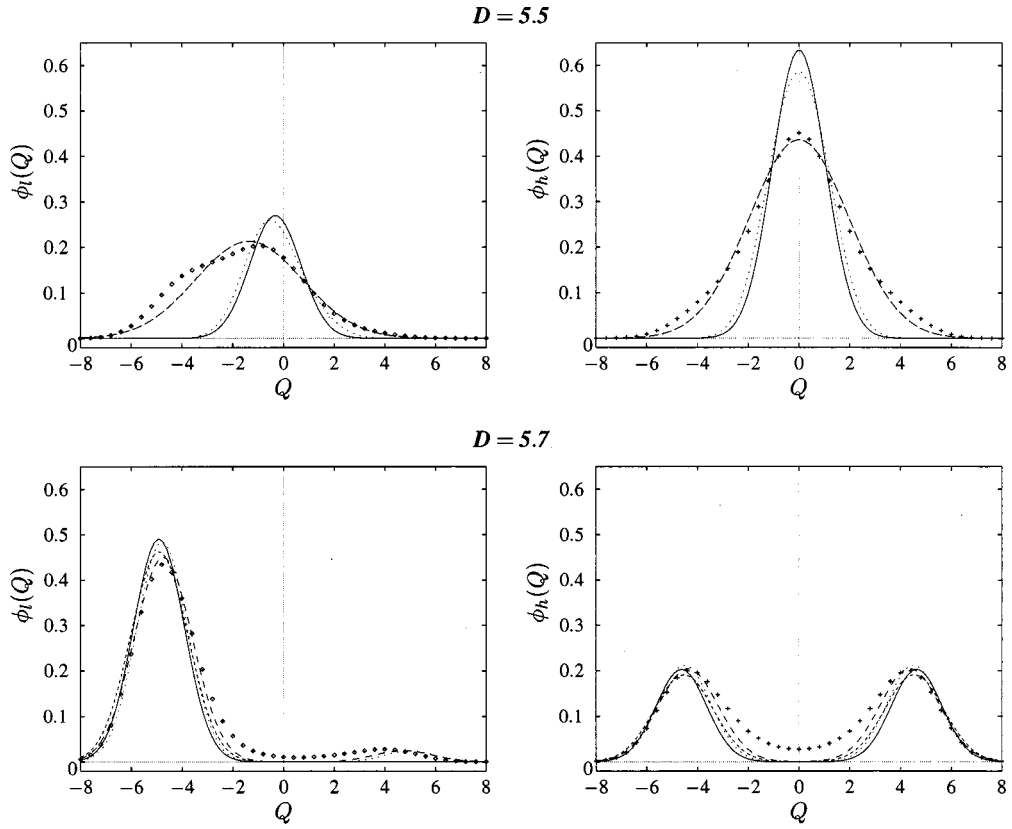
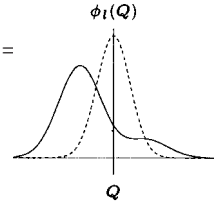


FIG. 15. Hubbard-bipolaron model ($U \neq 0$). Phononic partner ground state wave functions $\phi_l(Q)$ and $\phi_h(Q)$ [see Eq. (14)] for the electron-phonon coupling parameters $D=3.5$ and $D=4.2$ for $U=12.6$ and $T=10.1$: displacive ansatz (——), squeezing-displacive ansatz (- - -), nonunitary reflective ansatz (— — —), and unitary reflective-displacive ansatz (· · · ·). The exact result from matrix diagonalization is show by dots ($\diamond$ /+).

3. Nonunitary reflective ansatz

The nonunitary reflective ansatz is as follows:

$$\phi_l(Q) = \frac{(e^{i\delta_l P} + \eta \cdot e^{-i\delta_l P})}{\sqrt{1+\eta^2+2\eta \cdot \exp(-\delta_h^2 \cdot e^{4\gamma_l})}} e^{i\gamma_l(PQ+QP)} \varphi_0(Q) = \tag{43}$$

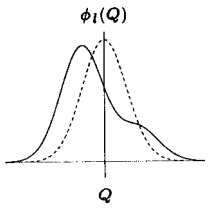


$\phi_h(Q)$: as in Eq. (42).

4. Unitary reflective-displacive ansatz

The unitary reflective-displacive ansatz is as follows:

$$\phi_l(Q) = e^{\tau Q R_Q} e^{i\delta_l P} \varphi_0(Q) = \tag{44}$$



$\phi_h(Q)$: as in Eq. (42)

C. Results

Figures 8 and 14 show the energetic behavior of the ground state for the Holstein ($U=0$) and Hubbard ($U\neq 0$) model, respectively. We observe that in both cases the nonunitary reflective ansatz (43) turns out to be the most favorable one and reproduces the exact numerical results in a quite satisfactory manner over the whole coupling range. By contrast, the squeezing-displacive ansatz (41), which frequently has been used in literature, is less good, and particularly in the high coupling regime it produces energy values which quite strongly derivate from the numerically exact ones. Comparing Fig. 8 and 14 we also note the more abrupt turnover to localization in the Hubbard case.

From Fig. 9 we note that only for the optimized nonunitary reflective ansatz and the squeezing displacive ansatz the displacement parameters display a somewhat smooth transition behavior in the turnover regime, as suggested by the $\langle Q^2\rangle$ ascent in Fig. 6.

From Figs. 10 and 13 we realize the decisive shortcoming of the squeezing-displacive ansatz (41) in the strong coupling regime, which is the abrupt return of $\phi_l(Q)$ to a nonsqueezed functional form (see lower left part of Fig. 13), shifted to the left side, taking no account of the small mirror-image peak on the right side, which is manifest in the exact numerical solution, and which is reproduced fairly adequate by the nonunitary reflective ansatz (43).

Figure 11 contains further information about the optimization of expressions (43) and (44) and Fig. 12 demonstrates again the superiority of the nonunitary reflective ansatz, yielding a smooth behavior in the turnover region. Figure 15 is the Hubbard-model ($U\neq 0$) equivalent of Fig. 13 and displays the same qualitative features as the latter.

V. SUMMARY AND PERSPECTIVES

In the presented work the 222 model, which is the most prominent of the bipolaronic prototype models, has been investigated. This model has been treated by several authors in various approximations. The aim of our work has been to establish and discuss analytical methods of approximations, which on the one hand improve the approximative situation and on the other hand display an option for generalizations to more complicated models. To this end we have exploited a symmetry adapted (Wigner) form for the wave function ($p=\pm 1$):

$$|\Psi_p\rangle=\left[\frac{1}{\sqrt{2}}\Big(|l\rangle\,\phi_l(Q)+p\cdot|r\rangle\cdot\underbrace{R_Q\phi_l(Q)}_{=\phi_r(Q)}\Big)+\frac{1}{2}(1+p)\cdot|h\rangle\,\phi_h(Q)\right] \tag{45}$$

where $\{|l\rangle,|r\rangle,|h\rangle\}$ is the two-electronic singlet base. This amounts to a unitary transformation (Fulton-Gouterman transformation) and allows for a generalization to more extended systems.[28] The Fulton-Gouterman transformation diagonalizes the Hamiltonian with respect to the electronic subsystem and establishes equations (''Fulton-Gouterman equations'') for the vibrational companion functions $\{\phi_l^{(p)}(Q),\phi_h^{(p)}(Q)\}$. These equations are approximately solved by means of optimized trial functions. The latter are generated by means of unitary operators (''displacive,'' ''squeezing,'' ''reflective,'' and combinations thereof). The considered functional forms for $\phi_l^{(p)}(Q)$ and $\phi_h^{(p)}(Q)$ qualitatively differ in their spatial behavior and, respectively, are compared with those of the exact solution given in Sec. II.

The most noticeable result is that by far the best ansatz for the ground state wave functions $\phi_l^{(g)}(Q)$ and $\phi_h^{(g)}(Q)$ is a reflective superposition of a deformed oscillatory function, which is generated by the application of a product of a squeezing and a displacive operator onto the undisturbed oscillatory function (''nonunitary reflective ansatz''). In particular it is to be emphasized that a combination of squeezing (or antisqueezing) and displacing (''squeezing-displacive ansatz''), which frequently has been employed in literature, is not *suitable for strong coupling* (see Fig. 8 and 14). The oscillatory companion functions $\phi_l^{(g)}(Q)$ and $\phi_h^{(g)}(Q)$ are bound to display a two-peaked nature (or at least a shoulder on one or both sides of the peak). This is a clear outcome of the exact numerical calculation (see Fig. 3 and 5). This finding has been reported already in the paper of Sander and Shore[9] in the one-electron model and discussed in detail in papers of us.[13,32] As regards squeezing, for the one-electron model it has been shown to be relevant only for highly excited states (''exotic states,'' see Refs. 33–35).

The main virtue of employing and studying unitary transformation for the generation of bipolaronic wave functions is the perspective that they lend themselves to generalization for more extended models which numerically cannot be handled. For example, the Fulton-Gouterman transformation can be generalized to systems with translational symmetry, as shown by one of us.[28] Also, the unitary operators used as generators for the phononic wave functions offer themselves to generalization.

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