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From electron to small polaron: An exact cluster solution

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We perform the exact diagonalization of two, four, and six vibrating molecules strongly coupled with one electron to show that a small polaron describes both adiabatic and nonadiabatic regimes. Electron spectra are derived and compared with the adiabatic Holstein approximation and with the Lang-Firsov canonical transformation in a wide range of the adiabatic parameter and the coupling strength. We show that the self-consistent Migdal approximation is violated at an intermediate value of the coupling constant $\lambda \approx 1$ and the adiabatic or nonadiabatic small polaron is the ground state for the intermediate and strong coupling $\lambda > 1$. The phonon frequencies are calculated for a wide range of the coupling, including the intermediate region.

I. INTRODUCTION

It has been known for a long time¹ that a system of electrons and phonons coupled by the Frohlich interaction is unstable at some critical value of the coupling. The phonon-vacuum instability appears in the adiabatic description¹ as an instability of bare phonons at $\lambda = \frac{1}{2}$: the renormalized phonon frequency $\tilde{\omega}$ being $\omega\sqrt{1-2\lambda}$.

The nature of the ground state in the strong-coupling limit $\lambda > 1$ is of great interest. The many-electron system on a lattice strongly coupled with any bosonic field turns out to be a charged Bose liquid (BL) consisting of on-site or intersite small bipolarons (charge $2e$, spin 0 or 1) if the Coulomb repulsion is not very large² or a heavy polaronic Fermi liquid in the opposite case.³ The point is that $\lambda \approx 1$ is the condition for the small polaron formation,⁴ which has been known for a long time⁵⁻⁹ as a solution of a single-fermion problem on a vibrating lattice. Once small polarons are formed the adiabatic approximation no longer holds because the electron band (no matter how wide it is) shrinks into a very narrow polaronic band.

The important point is that phonons are stable and their frequency renormalization is small (as $1/\lambda^2$) in the polaronic region of coupling, $\lambda \gg 1$, as has been shown with $1/\lambda$ expansion.¹⁰ Thus, the Frohlich Hamiltonian can be applied for any value of λ .

These results have been derived for a sufficiently large value of λ . Several attempts to describe the intermediate region of the coupling $\lambda \approx 1$ with and without electron-electron correlations are known in the literature. These are based on the variational approach,¹¹⁻¹⁴ the Monte Carlo calculations,^{15,16} and on the exact (numerical) solution of a several-site model.^{17,18} The general conclusion is that there is a continuous smooth evolution from a wide-band Fermi liquid to narrow-band small polarons (or bipolarons) in the intermediate region of the coupling $\lambda \approx 1$ and that neither Migdal nor small polaron approximations are in quantitative agreement with the exact re-

sult at intermediate-coupling strengths.

However, the numerical solutions of a several-site models have been given for a fixed value of the adiabatic parameter $\omega/t = 1.0$ (Ref. 17) or $\omega/t = 0.5$.¹⁸ At the same time these results have been compared with the Lang-Firsov approach, which is, strictly speaking, applicable only in the nonadiabatic limit $\omega/t > 1$. No comparison with the Holstein adiabatic small polaron has been made. Therefore, the applicability of the relevant small polaron theory in the intermediate-coupling region remains to be checked.

A numerical exact calculation of a self-trapped exciton was reported by Kongeter and Wagner¹⁹ for a different value of the adiabatic parameter. Two separated peaks in the spectral function were found and the disappearance of the second peak in the extreme nonadiabatic situation was stated.

In this paper we solve a model of two, four, and six vibrating molecules coupled with one electron in the adiabatic $\omega/t < 1$ as well as in the nonadiabatic $\omega/t > 1$ regimes using numerical diagonalization with 50 phonons for two sites and with 30 phonons for four and six sites. We show that the adiabatic Holstein small polaron and the Lang-Firsov canonical transformation are in excellent agreement with the exact solution in adiabatic and nonadiabatic regimes, respectively, for all values of the coupling strength. The Migdal approximation is only valid for a weakly coupled adiabatic system $\lambda, \omega/t \ll 1$. The phonon frequency renormalization is studied in a wide range of the coupling, including the intermediate region.

II. ANALYTICAL APPROACHES TO A STRONGLY COUPLED ELECTRON-PHONON SYSTEM

The familiar Frohlich Hamiltonian in the site representation has the following form:

FIG. 2. Ground-state energy of the extended cluster. (a) Four-site cluster for different types of boundary conditions. (b)–(d) Six-site cluster with periodic boundary condition.

III. NUMERICAL SOLUTION OF SEVERAL-SITE MODEL, COMPARISON WITH ANALYTICAL RESULTS

Two analytical approaches discussed above are based on the $1/\lambda$ or ω/t expansions. *A priori* it is difficult to judge to what extent they are reliable in the intermediate-coupling region, $\lambda \approx 1$ or for the intermediate value of the adiabatic parameter $\omega/t \approx 1$. It is clear that the ground-state energy as well as the renormalized phonon frequency are less sensitive to the parameters because they are the same in both approximations. However, the bandwidth might depend strongly on the approximation used. Fortunately, as we show below, analytical expressions, Eqs. (2) and (7) works perfectly well in the whole region $\lambda > 1$ for the nonadiabatic $\omega/t > 1$ and adiabatic $\omega/t < 1$ systems, respectively. This is in contrast with the conclusion¹⁸ based on the comparison of the numerical solution for the ground-state energy of an 8 and 16 molecular model in the *adiabatic* limit with the *nonadiabatic* Lang-Firsov formulas. Moreover, the corrections to the ground-state energy due to phonon frequency renormalization have not been taken into account in Ref. 18. These corrections are important in the intermediate-coupling limit $\lambda \sim 1$ and in the antiadiabatic limit $\omega > t$ due to zero-point fluctuations.

We present here the solution of two-, four-, and six-site models for a wide range of the adiabatic parameter ω/t . For $\omega/t = 1$ the two-site problem has been solved by Ranninger and Thiblin.¹⁷

After rewriting the Hamiltonian, Eq. (1), in terms of the modes corresponding to irreducible representations of the symmetry group of the linear chain a symmetric mode $x_0 = (u_1 + \dots + u_N)/\sqrt{N}$ couples with the constant total density of electrons $n = n_1 + \dots + n_N$ and can

be taken into account exactly (displaced oscillator). As a result the total number of phonon modes coupled with the electron is reduced to $N - 1$, $N = 2, 4$, or 6 . In the numerical calculations we take 50 phonons for $N = 2$, 10 phonons per each mode for $N = 4$, and 6 phonons per mode for $N = 6$.

To compare the exact results for a finite cluster with the adiabatic approximation we have solved a set of the adiabatic equations [Eqs. (3) and (4) in Ref. 22] for a finite cluster with one-loop correction which gives renormal-

FIG. 3. Renormalized phonon frequencies for a four-site cluster. All four phonon modes are shown, a bare asymmetric mode $(u_1, u_2, -u_2, -u_1)$ is adiabatically unstable at $\lambda \approx 0.57$.

ized phonon frequencies. The ground-state energy in the adiabatic approximation is

$$E_0 = \varepsilon_0 + 1/2 \sum_i^N \tilde{\omega}_i, \quad (10)$$

where $\tilde{\omega}_i$ is determined in terms of the solution of self-consistent equation²²

$$\text{Det}[\tilde{\omega}^2 \delta_{i,j} - \Pi_{i,j}(\tilde{\omega})] = 0, \quad (11)$$

where polarization is

$$\Pi_{i,j}(\Omega) = ig^2 \omega^2 \sum_{\omega} G_{i,j}(\omega) G_{j,i}(\Omega + \omega), \quad (12)$$

FIG. 4. Electronic spectral function for two-site (a) and (b) and four-site (c)–(f) models.

FIG. 5. The adiabatic terms of a two-site model. The upper curve corresponds to the nonbonding state.

$$G_{ij}(\omega) = \sum_n \frac{\psi_n(i)\psi_n(j)}{\omega - \epsilon_n}. \quad (13)$$

$\psi_n(i)$ is a wave function in the presence of polaronic potential well.

It should be noted that our approach is the same as an unrestricted symmetry-broken Hartree-Fock approximation.²⁹ The only difference from Ref. 29 is a different way of considering loop corrections. In our approach polarization loop has smaller matrix. Instead of the diagonalization of a bigger random-phase-approximation (RPA) ma-

trix²⁹ we solve the self-consistent equation (11).

The results of comparison of the adiabatic approximation with one-loop corrections and exact results are presented in Figs. 2(a)–2(d). As can be seen from Fig. 2(a) for $\omega=0.25t$ there is an excellent agreement of the ground-state energy in the adiabatic approximation with exact results in the whole range of the coupling strength. If the phonon frequency increases, the deviation of the ground-state energy in the adiabatic approximation from exact results appears in the intermediate-coupling range $\lambda \sim 1$. This deviation is connected with the fact that the renormalized phonon frequency is close to zero and the one-loop approximation is not sufficient. One can easily see this from the two-site model. For $\lambda < 1$ the renormalized phonon frequency is

$$\bar{\omega} = \omega \sqrt{1 - \lambda}. \quad (14)$$

In the region of $\lambda \sim 1$ the phonon frequency is small and the harmonic approximation for the adiabatic potential breaks down. The anharmonic effects are strong³⁰ and it is necessary to take into account anharmonic terms in the adiabatic potential.

The behavior of the local phonon modes as a function of the coupling constant λ for the four-site model is shown in Fig. 3. In the vicinity of $\lambda \sim 1$ the local phonon mode is close to the instability.

In the intermediate $\omega=t$ regime the ground-state energy for $\lambda > 1$ in the adiabatic approximation is also in good agreement with the exact results [Fig. 2(c)].

In the nonadiabatic limit $\omega=3t$, Fig. 2(d), we found good agreement of the Lang-Firsov approach after taking account of the phonon frequency renormalization (4) with exact results. It should be noted that there is no visible difference in the ground-state energy of the four-

FIG. 6. Phonon wave function: (a) $\omega/t=0.17$, (b) $\omega/t=0.43$, (c) $\omega/t=0.909$, (d) $\omega/t=3$.

site model with different types of boundary conditions (free ends and periodic boundary conditions) for $\lambda > 1$ [Fig. 2(a)]. This is due to the fact that the size of the small polaron is restricted to the lattice constant (see Refs. 7, 21, and 22) and the size of the system has no importance in that case.

Figures 4(a)–4(f) show the spectral function of two- and four-site models which correspond to the probability of absorption of a free electron with the momentum $\mathbf{k}$ into the ground state without any electrons as measured by the angle-resolved inverse photoemission spectroscopy.

$$A_{k,\sigma}(\epsilon) = 2\pi \sum_m \langle 0,0 | c_{k,\sigma} | m,1 \rangle \langle m,1 | c_{k\sigma}^\dagger | 0,0 \rangle \times \delta(\epsilon - E_m^1 + E_0^0),$$

where $|m,n\rangle$ and E_m^n are the eigenfunctions and eigenvalues of the Hamiltonian (1) with n electrons. Figures 4(a)–4(d) correspond to the adiabatic limit. In that case the spectral function has two well-pronounced maxima for the two-site model and four maxima for the four-site cluster. The first one corresponds to the polaronic phonon cloud. The states inside of this peak are responsible for the polaron formation. Other peaks appear due to the existence of the excited nonbonding states, which correspond to the upper adiabatic terms (Fig. 5). This peak is connected with the high-lying excited states ($E > g^2\omega$) in the polaronic potential well. In the adiabatic approximation these states are responsible for the midinfrared peaks in the optical conductivity.^{24–28} It should be noted that the existence of the second peak in the adiabatic limit has been reported earlier by Kongeter and Wagner.¹⁹

As can be seen from the Figs. 4(e) and 4(f) these non-

bonding states also exist in the intermediate regime ($\omega \sim t$) in contrast with the Monte Carlo simulations.¹⁵ In contrast to the pure adiabatic picture in the intermediate regime $\omega \sim t$ the polaron cloud and nonbonding states are close to each other and there is mixture of these states. In the antiadiabatic limit ($\omega > t$) the adiabatic potential does not exist due to fast phonon fluctuations and the second peak in the spectral function disappears as has been reported earlier.¹⁹

As can be seen from Figs. 4(c)–4(f) each line in the spectral function split into a few lines and these lines have different intensities. These peculiarities of the spectral function clearly show that phonon modes are not degenerate for this case due to a different renormalization factor (see also Fig. 3).

The calculated exact phonon wave function of a two-site model compared with the analytical one in the adiabatic approximation,

$$\Phi(X) = \left[\frac{1+\beta}{2} \right]^{1/2} \exp \left[-\frac{\tilde{\omega}(X-g\beta)^2}{2\omega} \right] + \left[\frac{1-\beta}{2} \right]^{1/2} \exp \left[-\frac{\tilde{\omega}(X+g\beta)^2}{2\omega} \right], \quad (15)$$

is shown in Figs. 6(a)–6(d). In contrast with Ranninger and Thibblin results¹⁷ we found excellent agreement of the phonon function with the adiabatic approximation up to $\omega \sim t$ [Fig. 6(c)]. The adiabatic approximation also gives a good description for the well-developed shoulder for $X < 0$. It should be noted that in the adiabatic limit the deformation is closely linked with the electronic wave

FIG. 7. Lowest-energy levels of a two-site model.

FIG. 8. "Bandwidth" of a two-site model.

function [see Eq. (11)] and a slow polaron tunneling corresponds to the tunneling of both deformation and electronic wave function. The shoulder of the phonon wave function is proportional to the density of the electron wave function on the neighboring site in the broken-symmetry ground state.

This shoulder is extremely important in the adiabatic limit when the phonon fluctuations are slow and the ground-state energy strongly depends on the form of the adiabatic potential.

In the antiadiabatic case this shoulder [Fig. 6(d)] becomes smaller and is not so important for the ground-state energy. This is connected with the fact that lattice fluctuations are fast and deformation relaxes quickly following the slow electronic motion. In that case there is good agreement with the Lang-Firsov nonadiabatic theory.¹⁷

Figures 7(a)–7(d) represent the result of the comparison of the exact energies of the first six levels with the small polaron levels, Eq. (6). As follows from the results there is an excellent agreement of the adiabatic theory with exact results for $\lambda > 1$ even in the intermediate regime $\omega \sim t$.

It should be noted that polaronic levels Eq. (6) in the adiabatic limit are degenerate. There are two equivalent positions of the polaron (sites 1 and 2) which correspond to the same energy. The splitting of these degenerate levels appears if we take into account tunneling processes. This splitting is proportional to the polaronic bandwidth.

Finally to analyze the polaronic bandwidth we calculate the splitting of two lowest levels. In that case we have two well-controlled approximations for the bandwidth, Eqs. (2) and (7). As can be seen from Figs. 8(a)–8(d) for $\omega = 0.1t$ there is a good agreement of exact results

with Eq. (7). It should be noted that the Lang-Firsov approximation gives in that case 2 orders-of-magnitude smaller value of the splitting. It means that in the adiabatic limit the procedure of averaging over phonons to estimate the polaronic bandwidth is incorrect. In the intermediate regime $\omega = 0.909t$ both formulas (2) and (7) give more or less a good estimate for splitting [Fig. 8(c)]. In the nonadiabatic limit [Fig. 8(d)] the energy splitting is in good agreement with the Lang-Firsov theory.

IV. CONCLUSION

Our conclusions are the following.

(i) The Lang-Firsov approach and the adiabatic Holstein approximation are in the excellent agreement with the exact diagonalization for the nonadiabatic $\omega/t > 1$ and adiabatic $\omega/t < 1$ system, respectively. We have also found that in the intermediate regime ($\omega \sim t$) both theories give a satisfactory estimate for the ground-state energy and the polaronic bandwidth.

(ii) In the intermediate-coupling region, $\lambda \simeq 1$, the Migdal approach is meaningless and the effective bandwidth of a small adiabatic polaron can be several orders of magnitude larger than that obtained with the nonadiabatic formula, Fig. 8. This drastically reduces its effective mass.

(iii) We have found that the spectral function has two separated peaks in the adiabatic limit (Fig. 4), as has been reported earlier.¹⁹ We also found that second high-energy peak disappears in the nonadiabatic limit ($\omega > t$) due to mixture with a polaronic cloud.¹⁹

(iv) In the intermediate-coupling regime ($\lambda \sim 1$) in the adiabatic limit where a double-well structure appears in the adiabatic potential and renormalized phonon frequen-

cy is close to zero (Fig. 3) anharmonic effects are important.³¹

(v) The renormalized phonon frequencies are found, Fig. 3, in a wide range of the coupling, including the intermediate region.

It is necessary to mention that we have restricted our analysis to the case of one electron or one hole and consequently the Coulomb repulsion is absent from our Hamiltonian Eq. (1).

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APPENDIX

The electron self-energy Σ in the Migdal approximation contains two contributions, Σ_M and Σ_μ , Fig. 9. $\Sigma_M \simeq \lambda\omega$ and thus remains small compared with the bandwidth $\simeq t$ in the relevant region of the coupling ($\lambda < t/\omega$), which guarantees the self-consistency of the approximation. On the other hand, $\Sigma_\mu = -g^2\omega n$ turns out to be comparable to or larger than the Fermi energy already at $\lambda > 1$ for any filling of the band, as it was mentioned in Ref. 32, where n is the electron density.

As a rule this diagram, which is momentum and frequency independent, is included in the definition of the chemical potential μ . However, this is justified only in the weak-coupling regime. For a strong coupling $\lambda > 1$

FIG. 9. Electron self-energy in the Migdal approximation.

Σ_μ leads to an instability. To show this let us consider a one-dimensional chain. The renormalized chemical potential is given by

$$\mu = 2t \sin \left[\frac{\pi(n-1)}{2} \right] - 2t\lambda n. \quad (\text{A1})$$

The system is stable if $d\mu/dn$ is positive, which yields the following region of the stability of the Migdal solution:

$$\lambda < \frac{\pi}{2} \cos \left[\frac{\pi(n-1)}{2} \right]. \quad (\text{A2})$$

For two- and three-dimensional lattices the numerical coefficient is different, but the critical value of λ remains to be of the order of unity. This consideration unambiguously shows that the extension of the Migdal approximation to the strong-coupling region $\lambda > 1$ is unacceptable. For a filling different from $\frac{1}{2}$ the critical value of λ above which the Migdal approximation is violated turns to be even less than unity.

It is important that in the 1D case the polaron forms for any strength of the coupling.¹¹ This is clearly seen from Eq. (A2). For $n \rightarrow 0$ the critical value λ_c is equal to 0. It means that system is unstable with respect to formation of localized polarons. Delocalization of the polaron takes place due to tunneling processes, which are nonperturbative.

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⁴The critical value of λ depends on the type of the electron-phonon interaction. In simple metals like Pb the crystal field is screened, and the phonon-assisted intersite hopping dominates in the electron-phonon interaction. This hopping can destroy polarons. That is why Pb, with $\lambda > 1$, shows no sign of polarons.

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