Polaron band formation in the Holstein model

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We present numerical exact results for the polaronic band structure of the Holstein molecular crystal model in one and two dimensions. The use of a direct Lanczos diagonalization technique, preserving the full dynamics and quantum nature of phonons, allows us to analyze in detail the renormalization of both quasiparticle bandwidth and dispersion by the electron-phonon interaction. For the two-dimensional case some of our exact data are compared with the results obtained in the framework of a recently developed finite-cluster strong-coupling perturbation theory. [S0163-1829(97)04131-3]

The very fundamental problem of a single conduction electron coupled to bosonic degrees of freedom is still not completely understood. In the case where the bosons are lattice phonons that is what is usually called the polaron problem and numerous analytical techniques were tried out to tackle this complicated many-body problem in terms of the simplest electron-phonon (EP) Hamiltonians.[1] Depending on the relative importance of the long-range or short-range electron-lattice interactions simplified models of the Fröhlich[2] or Holstein[3] type, respectively, have been widely used to analyze polaronic effects in solids with displaceable atoms. The crucial point is that the usual phase transition concept fails to describe the crossover from an only weakly dressed electron to the strongly renormalized less-mobile polaronic quasiparticle with increasing EP coupling strength. Furthermore, as yet none of the various analytical treatments, based on variational approaches[4] or on weak-coupling[5] and strong-coupling adiabatic[3] and nonadiabatic[6,7] perturbation expansions, are suitable for the investigation of the physically most interesting transition region, where the highly nonlinear ''self-trapping'' process of the charge carrier takes place. That is because precisely in this situation the characteristic electronic and phononic energy scales are not well separated and nonadiabatic effects become increasingly important, implying a breakdown of the standard Migdal approximation.[8] In principle, quasi-approximation-free numerical methods like (quantum) Monte Carlo simulations[9] and exact diagonalizations[10] can close the gap between the weak and strong EP coupling limits and therefore, at the moment, provide the only reliable tool for studying polarons close to the crossover regime. Along this line, previous exact diagonalization work has concentrated on spectral properties of the one-dimensional (1D) Holstein polaron. In practice, however, memory limitations have restricted the calculations to either small values of the EP coupling constant or to very small clusters,[11–13] thus, e.g., making a more detailed discussion of the polaronic band dispersion impossible. First exact diagonalization studies performed on the 1D ten-site Holstein model indicate that the small polaron dispersion may differ significantly from a rescaled cosine tight-binding band in the crossover regime;[14] a result which has been corroborated more recently by a finite-cluster strong-coupling perturbation theory (SCPT).[15]

Encouraged by these findings, it is the aim of the present paper to perform a systematic Lanczos study of the 1D and 2D Holstein model on large enough lattices, in order to discuss the evolution of the polaronic quasiparticle band structure in dependence on the phonon frequency and EP interaction strength.

The Holstein Hamiltonian reads

$$\mathcal{H}=-t\sum_{\langle ij\rangle}(c_i^\dagger c_j+c_j^\dagger c_i)-\sqrt{\varepsilon_p\hbar\omega}\sum_i(b_i^\dagger+b_i)c_i^\dagger c_i+\hbar\omega\sum_i\left(b_i^\dagger b_i+\frac{1}{2}\right), \tag{1}$$

where $c_i^{[\dagger]}$ and $b_i^{[\dagger]}$ are the annihilation [creation] operators of a (spinless) fermion and a boson (phonon) at Wannier site i, respectively. In Eq. (1), the following idealizations of real electron-phonon systems are made: (i) the electron transfer t is restricted to *nearest-neighbor* (NN) pairs $\langle ij\rangle$; (ii) the charge carrier is *locally* coupled to a *dispersionless* optical phonon mode (ε_p denotes the EP coupling constant and $\hbar\omega$ is the bare phonon frequency); (iii) the phonons are treated within an *harmonic* approximation. Recall that the physics of the Holstein model is governed by *two* dimensionless parameters $\lambda=\varepsilon_p/2Dt$ and $\alpha=\sqrt{\varepsilon_p/\hbar\omega}$; i.e., irrespective of the adiabaticity ratio $\gamma=t/\hbar\omega$, small polarons are formed provided that $\lambda>1$ *and* $\alpha>1$.

In the numerical analysis of the Holstein model, we employ the standard Lanczos algorithm in combination with a well-controlled truncation of the phononic Hilbert space. A general state of the Holstein Hamiltonian is given as the direct product $|\Psi\rangle=\Sigma_{r,s}c_r^s\{|r\rangle_{\rm el}\otimes|s\rangle_{\rm ph}\}$ with $|s\rangle_{\rm ph}=\Pi_{i=1}^N(1/\sqrt{n_i^s!})(b_i^\dagger)^{n_i^s}|0\rangle_{\rm ph}$, where r and s label the basis states of the electronic and phononic Hilbert space with dimensions $D_{\rm el}=N$ and $D_{\rm ph}$, respectively, i.e., $D_{\rm tot}=D_{\rm el}\times D_{\rm ph}$. Restricting ourselves to at most M phonons on a finite lattice with N sites we take into account all m-phonon states $m=\Sigma_{i=1}^N n_i^s\leqslant M$ with $n_i^s\in[0,m]$ and $1\leqslant s\leqslant D_{\rm ph}^{(M)}=(M+N)!/M!N!$. To further reduce the dimension of the Hamiltonian matrix to be diagonalized, we exploit the translational invariance of our finite system by the use of symmetrized basis sets,

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$$\{|\vec{K},s\rangle\}=\frac{1}{\sqrt{N}}\sum_{i=1}^{N} e^{i\vec{K}\vec{R}_i}\mathcal{T}_{\vec{R}_i}(|1,0,\ldots,0\rangle\otimes\{|s\rangle_{\rm ph}\}). \quad (2)$$

In Eq. (2), $\mathcal{T}_{\vec{R}_i}$ describes an allowed lattice translation. Thus in each $\vec{K}$ subspace we have $D_{\rm tot,\vec{K}}=D_{\rm ph}^{(M)}$. Note that $\vec{K}$ is the *total* momentum of the coupled EP system. According to the phononic truncation procedure, which is described in detail in Ref. 14, we have to check for the convergence of both the ground-state energy $E_{0,\vec{K}}^{(1)}(M)$ [the superscript ''(1)'' refers to the single-particle case] and the phonon distribution function $|c^m|^2(M)=\Sigma_{r,s}|c_s^r|^2$. Increasing the maximum number of phonons retained (M), convergence is assumed to be achieved if both the relative error $|E_{0,\vec{K}}^{(1)}(M+1)-E_{0,\vec{K}}^{(1)}(M)|/|E_{0,\vec{K}}^{(1)}(M)|$ and $|c^M|^2$ become less than 10^{-7}.

In order to discuss the single-polaron polaron band structure one has to evaluate the wave-vector resolved spectral density function,

$$A_{\vec{K}}(E)=\sum_n |\langle\Psi_{n,\vec{K}}^{(1)}|c_{\vec{K}}^\dagger|0\rangle|^2\,\delta(E-E_{n,\vec{K}}^{(1)}). \quad (3)$$

This can be done using popular techniques for calculating spectral properties of Hamiltonian matrices, such as the standard Lanczos recursion method. However, for the small polaron problem an extreme energy resolution of the lower part of the single-particle spectrum is required. Therefore we apply a more advanced algorithm, based on a combination of Chebyshev recursion and maximum entropy, which is very efficient and numerically stable for high-energy resolution applications.[16] Then the so-called ''coherent'' band dispersion, $E_{\vec{K}}$, is obtained from the first peak of $A_{\vec{K}}(E)$ having finite spectral weight,

$$Z_{\vec{K}}=|\langle\Psi_{0,\vec{K}}^{(1)}|c_{\vec{K}}^\dagger|0\rangle|^2, \quad (4)$$

where $|\Psi_{0,\vec{K}}^{(1)}\rangle$ denotes the single-polaron state being lowest in energy in a certain $\vec{K}$ sector.

In the first place, we focus on the 1D model and start with the study of the *weak-coupling case* ($\lambda<1$). Figure 1 displays the band dispersion calculated at $\varepsilon_p=0.1$ for various phonon frequencies ranging from the adiabatic ($\gamma=10$) to the nonadiabatic ($\gamma=0.125$) regime (in the following all energies are measured in units of t). For low and intermediate phonon frequencies, the energy to excite one phonon lies inside the bare tight-binding band, $\epsilon_{\vec{K}}=-2t\Sigma_{i=1}^{D}\cos K_i$, of a D-dimensional hypercubic lattice with unit lattice spacing. Thus at arbitrarily small ε_p, predominantly ''phononic'' states enter the low-energy spectrum in those $\vec{K}$ sectors for which $\epsilon_{\vec{K}}$ is separated from the ground state by an energy $E\gtrsim\hbar\omega$. As a result the dispersion curves reflect the well-known peculiarities of the absorption spectra of an electronic or excitonic system weakly interacting with dispersionless optical phonons,[17,18] i.e., a nearly unaffected cosine dispersion near the band center ($\vec{K}=0$) and a practically flat region at larger momenta. Note that even in cases where the phonon frequency is comparable to the bare electronic bandwidth a remarkable ''flattening'' of the band structure appears in the vicinity of the zone boundary [e.g., at $\hbar\omega=4$ we found $E_\pi(\varepsilon_p=0.1)/E_\pi(0)\sim 0.92$]. In the extreme nonadiabatic regime ($\gamma\ll 1$) the phonon distribution function is sharply peaked at the zero-phonon state and, because the interaction is weak and the phonons can follow the electron instantaneously, we obtain a negligible renormalization of the band structure. To substantiate this interpretation we have calculated the $\vec{K}$-dependent mean phonon number,

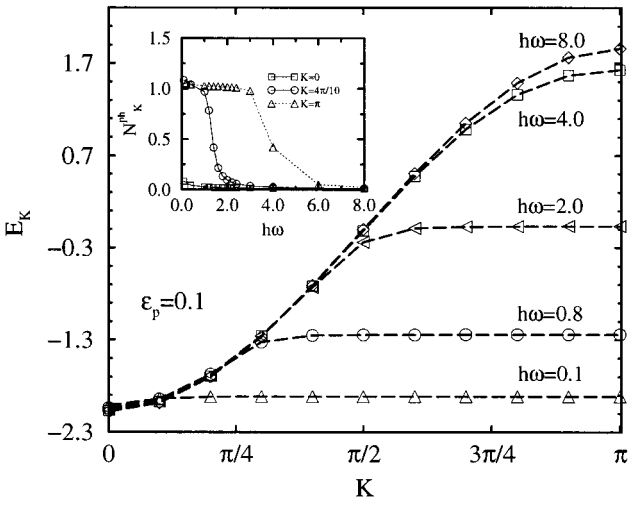

FIG. 1. Polaronic band dispersion $E_{\vec{K}}$ for the 1D Holstein model. Data points are exact results obtained for a finite lattice with $N=20$ sites and at most $M=10$ phonons using periodic boundary conditions. Here and in the following the energy scales of the ordinates were shifted by $N\hbar\omega/2$. The inset shows the mean phonon number $N_{\vec{K}}^{\rm ph}$ in band states with wave-vector $\vec{K}$.

$$N_{\vec{K}}^{ph}=\sum_i \langle\Psi_{0,\vec{K}}^{(1)}|b_i^\dagger b_i|\Psi_{0,\vec{K}}^{(1)}\rangle, \quad (5)$$

in the lowest band states $|\Psi_{0,\vec{K}}^{(1)}\rangle$. The results are presented in the inset of Fig. 1 for three characteristic momenta $\vec{K}=0,\ 0.4\pi$, and π. Evidently the states keeping small momenta are basically zero-phonon states. For larger momentum, we observe a gradual crossover from one-phonon to zero-phonon states upon increasing the phonon frequency, whereby the admixture of ''electronic character'' increases.

Of course it is of special interest to understand the evolution of the band structure by increasing the EP coupling strength. Thus it is fruitful to compare $E_{\vec{K}}$ for the weak and strong-coupling cases at a fixed phonon frequency. This has been done in Fig. 2 for various system sizes $N=10,\ldots,20$, using EP couplings $\varepsilon_p=0.5$ (a) and $\varepsilon_p=4.0$ (b) together with an intermediate adiabaticity parameter $\gamma=1.25$. First of all one notices that the finite-size effects are very small, i.e., although the data points belong to different lattice sizes we obtain a remarkably smooth behavior of $E_{\vec{K}}$. As might be expected, for $\varepsilon_p=0.5$ the coherent bandwidth, $\Delta E=\sup_{\vec{K}}E_{\vec{K}}-\inf_{\vec{K}}E_{\vec{K}}$, is approximately given by the phonon energy ($\Delta E=0.782\sim\hbar\omega=0.8$) and the dispersion again shows the coexistence of two different types of band states at small and large momenta. By contrast, if the EP interaction is enhanced, small polaron formation takes place and we observe all signs of the famous polaronic band collapse. However, note that even in the relatively strong EP coupling regime displayed Fig. 2(b) the standard Lang-Firsov formula, $\Delta E_{LF}=4Dt\exp[-\varepsilon_p/\hbar\omega]$ (obtained by performing the

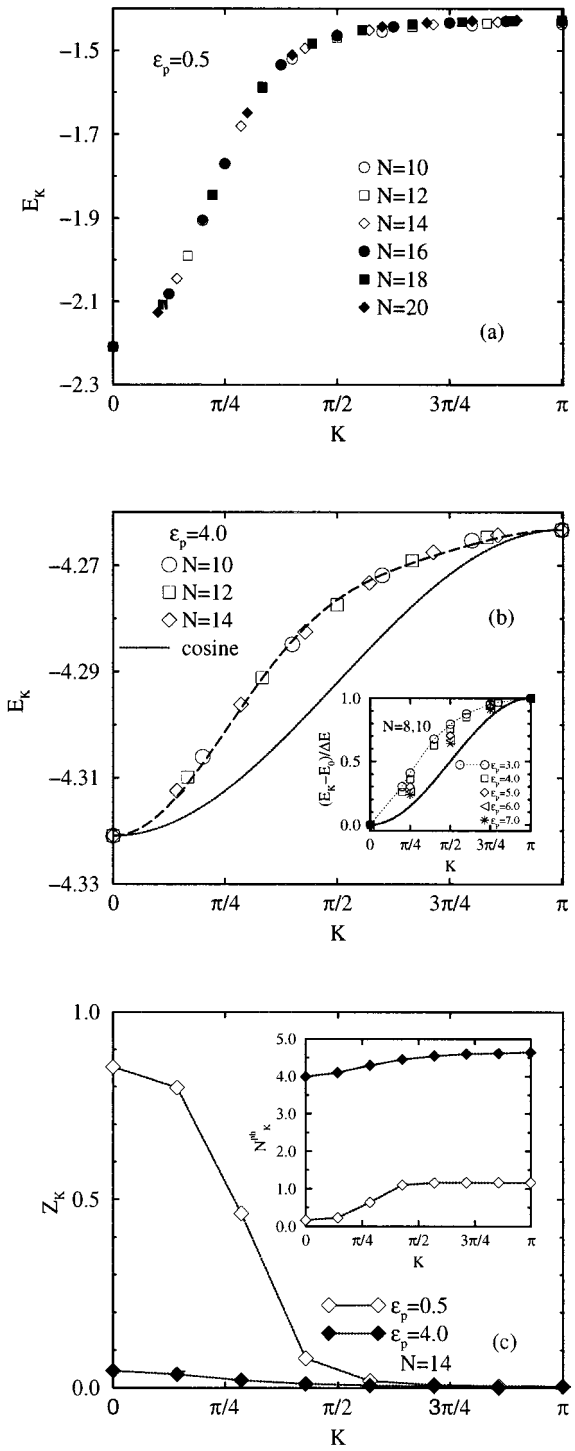

FIG. 2. Polaronic band structure of the 1D Holstein model in the weak (a) and strong (b) coupling regimes, where the phonon frequency is $\hbar\omega=0.8$. For both cases, the quasiparticle residue and mean phonon number are given in (c).

Lang-Firsov canonical transformation[6] and taking the expectation value of the kinetic energy over the transformed phonon vacuum), does not give a satisfactory estimate of the bandwidth. So we found $\Delta E_{\mathrm{LF}}=0.0269$ which has to be contrasted with the exact result $\Delta E=0.0579$. Besides the band narrowing effect, there are several other features worth mentioning for polaronic band states in the crossover region. Most notably, throughout the whole Brillouin zone the band structure differs significantly from that of a rescaled tight-binding (cosine) band containing only NN hopping. As can be seen from a least-squares fit to an effective band dispersion $\bar{E}_{\vec{K}}=\Sigma_{l=0}^{3}a_l\cos lK$, the residual polaron-phonon interaction generates *longer-range* hopping terms.[14,19] Concomitantly, the mass enhancement due to the EP interaction is weakened at the band minimum. It is important to realize that these effects are most pronounced at *intermediate* EP couplings and phonon frequencies. In this parameter region even higher-order SCPT, with its internal states containing some excited phonons, seems to be not tractable because the convergence of the series expansion is poor.[15] The deviation from the cosine dispersion at different ε_p is depicted in the inset of Fig. 2(b), where we have used the eight- and ten-site lattices in order to reach larger EP coupling strengths. Here $E_{\vec{K}}$ is scaled with respect to the coherent bandwidth which strongly depends on ε_p and $\hbar\omega$, for example, for $N=8$ and $\hbar\omega=0.8$ we found $\Delta E(\varepsilon_p)=0.1957$, 0.0559, 0.0143, 0.0037, and 0.0010 for $\varepsilon_p=3.0$, 4.0, 5.0, 6.0, and 7.0, respectively. It is obvious and does not need discussion that the dispersion is barely changed from a rescaled tight-binding band in the very extreme small polaron limit ($\lambda\gg1$). To visualize in more detail the different nature of large and small polaron band states, we have evaluated numerically the quasiparticle residue $Z_{\vec{K}}$ and the mean phonon number $N_{\vec{K}}^{(\mathrm{ph})}$ at all $\vec{K}$ points. Obviously, the $\vec{K}$-dependent renormalization factor can be taken as a measure of the ''electronic contribution'' to the polaronic quasiparticle. The results are shown in Fig. 2(c). For weak EP coupling, one recognizes immediately the electronic and phononic character of the states at small and large momentum, respectively. The crossover between these two types of states is accompanied by a change in the mean phonon number by approximately one. With increasing ε_p a strong mixing of electron and phonon degrees of freedom takes place, whereby, forming a small polaron, both quantum objects completely lose their own identity. As expected, this leads to a significant suppression of the quasiparticle residue for all $\vec{K}$. At large ε_p, the polaronic state is characterized by strong on-site electron-phonon correlations making the quasiparticle susceptible to self-trapping. Thus the coherent movement of the carrier is greatly hindered but itinerant band states do still exist. The small polaron is heavy because it has to drag with it a large number of phonons in the so-called ''phonon cloud'' [cf. Fig. 2(c) inset]. Since the high-energy band edge states are more vulnerable to decay from EP interaction the particle loses its spectral weight more rapidly at $\vec{K}=\pi$.[20]

Next, we consider the evolution of the band structure for the two-dimensional case. Figure 3 presents the results obtained for $E_{\vec{K}}$ and $Z_{\vec{K}}$ along the highly symmetric directions of the Brillouin zone in the weak- (a) and intermediate- (b) coupling regime. As in the (weak-coupling) 1D case, we found a flattening of the band structure in regions where $\epsilon_{\vec{K}}$ becomes comparable to the phonon energy. The sharp drop in the quasiparticle weight obtained with increasing $|\vec{K}|$ at small $\hbar\omega$ is weakened in the nonadiabatic regime. That is, the transition region ($0.25\lesssim Z_{\vec{K}}\lesssim0.75$) is enlarged. In order to make a comparison with recent predictions of both second-order Rayleigh-Schrödinger SCPT and the resummed finite-cluster SCPT developed by Stephan,[15] we have plotted

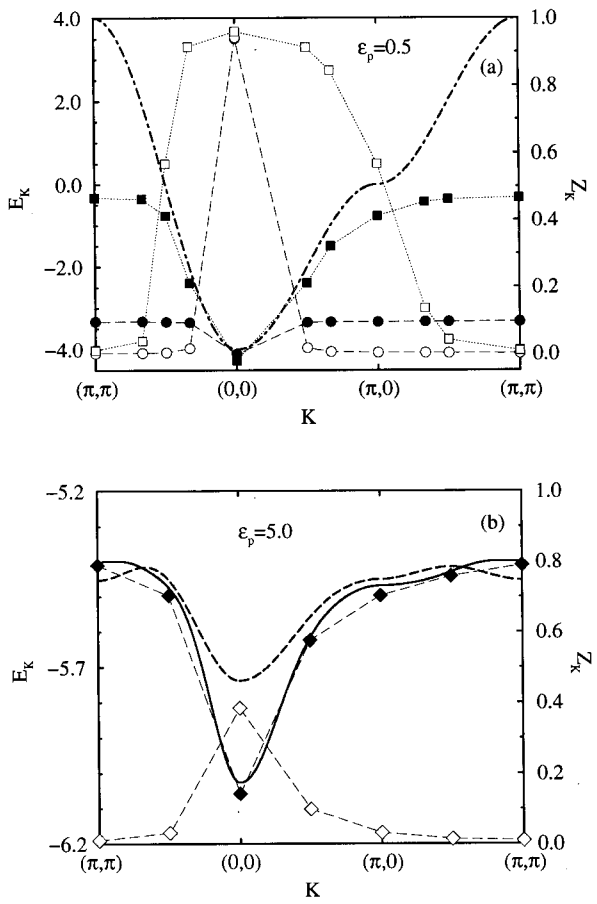

FIG. 3. Band dispersion $E_{\vec{K}}$ (filled symbols) and quasiparticle weight factor $Z_{\vec{K}}$ (open symbols) for the 2D Holstein model with $\varepsilon_p=0.5$, $\hbar\omega=0.8$ (circles), 4.0 (squares), and $\varepsilon_p=5.0$, $\hbar\omega=1.5$ (diamonds). Exact diagonalization results are obtained using finite square lattices with $N=16$ and 18 sites (the lines are only a guide for the eye). In the weak-coupling case (a) the chain dashed curve gives the free electron dispersion $\epsilon_{\vec{K}}$; for the strong-coupling case (b) exact data (filled diamonds) are compared with the predictions of standard second-order SCPT (dashed line) and finite-cluster SCPT (solid line) (Ref. 15).

$E_{\vec{K}}$ and $Z_{\vec{K}}$ for $\varepsilon_p=5.0$ and $\hbar\omega=1.5$ in Fig. 3(b). Interestingly enough, the flattening of the dispersion at the edges of the Brillouin zone survives to surprisingly strong EP couplings (note that $\hbar\omega\gtrsim 2\Delta E$). The wave-vector renormalization factor $Z_{\vec{K}}$ clearly indicates the polaronic nature of the charge carrier. That means, calculating the strong-coupling wave function we have to include multiphonon states. Once again the Lang-Firsov formula (first-order SCPT) underestimates the exact bandwidth by more than a factor of two; $\Delta E_{\mathrm{LF}}=0.2854$. Even if we include second-order corrections the SCPT band structure significantly deviates from the exact result. Obviously, the agreement is poorest at the Γ point ($\vec{K}=0$). Thus we are lead to the conclusion that the standard strong-coupling approximation fails to accurately predict the ground state energy and the polaronic bandwidth in the moderate strong-coupling regime. By contrast, the finite-cluster SCPT approach,[15] which in our case makes use of information extracted from a sequence of finite-cluster diagonalizations up to five sites, provides a correct description of the coherent band structure. The extremely good success of the finite-cluster SCPT at intermediate EP couplings and phonon frequencies may be attributed to the inclusion of longer-range correlations, which are especially important in the crossover region from nearly free to small polarons.

To summarize, we have performed a comprehensive study of the 1D and 2D Holstein model using exact diagonalizations. Concerning the polaronic band structure of the Holstein model, our main results are the following.

(i) In the weak-coupling case and for phonon frequencies less than the bare electronic bandwidth, we recover the expected flattening of the band dispersion at large momentum. Here our numerical results for the wave-vector resolved quasiparticle weight factor and the mean phonon number clearly demonstrate the electronic and phononic character of the band states near the band center and band edges, respectively.

(ii) Increasing the electron-phonon coupling the flattening survives to a large extent the crossover from large to small polarons. The small polaron state is characterized by less spectral weight and a large average number of phonons.

(iii) At intermediate EP coupling strengths and phonon frequencies the effective polaronic band dispersion deviates substantially from a simple tight-binding cosine band due to further than nearest-neighbor ranged hopping processes generated by the residual polaron-phonon interaction, implying the importance of multiphonon states. In the intermediate-coupling regime, the polaronic mass enhancement can be one order of magnitude smaller than predicted by standard strong-coupling perturbation theory.

(iv) In the extreme strong-coupling limit ($\varepsilon_p\gg\hbar\omega,t$), we have a nearly dispersionless small polaron band with almost vanishing spectral weight. That means, here the exponential band narrowing completely dominates the physics of the Holstein model and, in real solids, coherent polaron motion will be immediately destroyed by disorder effects.

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