

Effect of dispersive optical phonons on the behavior of a Holstein polaron

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We use the approximation-free bold diagrammatic Monte Carlo technique to study the effects of a finite dispersion of the optical phonon mode on the properties of the Holstein polaron, especially its effective mass. For weak electron-phonon coupling the effect is very small, but it becomes significant for moderate and large electron-phonon coupling. The effective mass is found to increase (decrease) if the phonon dispersion has a negative (positive) curvature at the center of the Brillouin zone.

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I. INTRODUCTION

Electron-phonon (e-ph) coupling and its effects on the properties of quasiparticles is one of the fundamental topics in condensed matter physics.¹ In particular, for a very weakly doped insulator, one can investigate the interactions of a single carrier with the phonon distortion that builds up in its presence to understand the properties of a single polaron: the dressed quasiparticle consisting of the carrier and its phonon cloud.

Most such polaron studies focus on e-ph coupling to optical phonons. The reason is that (assuming that long-range interactions are screened) the strength of the short-range coupling depends on the relative displacements between the atom hosting the carrier and its neighbors because these displacements modulate the onsite energy and hopping integrals of the carrier. Acoustic phonons are gapless and thus easy to excite, however, since they describe “in-phase” motion of neighboring atoms,¹ the corresponding relative displacements are vanishingly small leading to very weak e-ph coupling. In contrast, optical phonons describe “antiphase” atomic motion¹ which leads to large relative displacements and therefore a much stronger e-ph coupling.

Aside from ignoring coupling to acoustic phonons,² another widespread approximation is to assume that the optical phonon is dispersionless (Einstein model).³ In part, this is due to the belief that this should be a good approximation if the phonon bandwidth is small compared to the average phonon energy.⁴ In practice, this is also due to the scarcity of numerical techniques suitable for the study of polaron models with dispersive phonons. These reasons explain why very little is known about the effect of dispersive optical phonons on polaron properties. For example, all else being equal, will dispersive phonons increase or decrease the polaron’s mass? The fact that the phonons have a finite speed might suggest the former since the cloud is now mobile. However, mobile phonons will spread in all directions away from the carrier; this may lead to a more extended and thus harder to move cloud (for a fixed number of phonons). Apart from clarifying qualitative trends, it is important to understand quantitatively the accuracy of this approximation, as it may have important consequences in the modeling of materials and of quantum simulators.⁵

In the wake of the discovery of several instances where what were long believed to be standard polaronic properties were found to break down upon straightforward extensions of polaronic models,^{6,7} we believe it is high time to reexamine

this approximation of an Einstein mode.³ Here, we investigate the effect of dispersive phonons in the most well-understood polaron model with short-range e-ph coupling,⁸ the Holstein model.⁹

II. MODEL AND METHODS

We consider the generalized Holstein model for a one-dimensional chain with $N \rightarrow \infty$ sites and lattice constant $a = 1$:

$$\hat{\mathcal{H}} = \sum_k \epsilon(k) c_k^\dagger c_k + \sum_q \Omega(q) b_q^\dagger b_q + \frac{g}{\sqrt{N}} \sum_{k,q} c_{k+q}^\dagger c_k (b_{-q}^\dagger + b_q), \quad (1)$$

where c_k annihilates a carrier with momentum k (the spin degree of freedom is trivial so it is not listed) while b_q annihilates a phonon with momentum q . Sums are over the Brillouin zone $-\pi < k \leq \pi$. The first term is the carrier’s kinetic energy with the usual tight-binding dispersion $\epsilon(k) = -2t \cos(k)$; t is the hopping constant. The second term describes the phonons. For simplicity, we forego an exact description of the optical phonon dispersion for a detailed model of a lattice with a nontrivial basis, and instead use

$$\Omega(q) = \Omega_0 - \Delta\Omega/2[1 - \cos(q)]. \quad (2)$$

$\Omega_0 = \Omega(0)$ is the long-wavelength limit of the phonon energy. Since $\Omega(\pi) = \Omega_0 - \Delta\Omega$, the phonon bandwidth is $\Delta\Omega$. We will focus on downward (normal) dispersion with $0 < \Delta\Omega < \Omega_0$, but will also comment on the case with $\Delta\Omega < 0$. The third term is the Holstein coupling.⁹

Taken at face value, this Hamiltonian is unphysical: a chain with one atom per unit cell does not support optical phonons. One should think of the orbitals kept in the model as being the subset of valence orbitals associated with a more complex unit cell, with the assumption that the other orbitals are located at energies far enough removed from these that their occupation numbers can not change. From a pragmatic point of view, our motivation is to study a polaron model that is very well understood for dispersionless phonons, to see the effects of changing only this assumption.

The properties of the Holstein polaron for Einstein phonons ($\Delta\Omega = 0$) are controlled by two dimensionless parameters: (i) the effective coupling $\lambda = g^2/(zt\Omega_0)$ equal to the ratio of

the deformation energy $-g^2/\Omega_0$ (the polaron energy in the impurity limit $t = 0$) to the free electron energy $-zt$, z being the coordination number [$z = 2$ in one dimension (1D)]; for $\Omega(q)$ of Eq. (2) the deformation energy is $-g^2/\sqrt{\Omega_0(\Omega_0 - \Delta\Omega)}$ (see following), thus

$$\lambda = \frac{g^2}{2t\sqrt{\Omega_0(\Omega_0 - \Delta\Omega)}}, \quad (3)$$

and (ii) the adiabaticity ratio Ω_0/t . We round up these parameters with (iii) the dimensionless phonon bandwidth $\delta = \Delta\Omega/\Omega_0 < 1$. Hereafter, we set $t = 1$.

We investigate the effect of $\Delta\Omega \neq 0$ on the polaron energy $E(k)$, i.e., the energy of the lowest eigenstate of momentum k , $\hat{\mathcal{H}}|\vec{k}\rangle = E(k)|\vec{k}\rangle$, and its quasiparticle weight $Z(k) = |\langle k|\vec{k}\rangle|^2$ given by the overlap between the polaron eigenstate $|\vec{k}\rangle$ and the free-electron state $|k\rangle = c_k^\dagger|0\rangle$. We also study the effective polaron mass $m^*/m_0 = 2t/\frac{\partial^2 E(k)}{\partial k^2}|_{k=0}$, where the bare electron mass is $m_0 = 1/2t$.

The results presented here are obtained with a variant of the diagrammatic Monte Carlo (DMC) technique¹⁰⁻¹² known as the bold diagrammatic Monte Carlo (BDMC) technique.^{6,13} Like DMC, BDMC consists in a Monte Carlo sampling of the Feynman diagrammatic expansion of the continuous imaginary-time self-energy of the polaron. The difference is that this sampling is done with the electron bare propagator self-consistently replaced by a dressed propagator as the calculation progresses (thus speeding up convergence), while enforcing necessary restrictions on the topology of the diagrams to avoid double counting. As is the case for DMC, BDMC is exact within the limits of its statistical error bars and does not make any assumptions or enforce any nonphysical restrictions. DMC and its variants can treat dispersive (including acoustic) phonons efficiently, provided we avoid the extreme adiabatic regime $\Omega_0/t \ll 1$.

III. RESULTS AND DISCUSSION

Figure 1 shows BDMC results for $E(k)$ and $Z(k)$ for increasing $\Delta\Omega/\Omega_0$ and different values of λ . In all cases $t = 1$, $\Omega_0 = 2$. The error bars are smaller than the size of the symbols. Consider first $\Delta\Omega = 0$, i.e., the usual Holstein model. As expected,⁸ both $E(k)$ and $Z(k)$ are monotonic functions of the momentum k . With increasing λ , the polaron bandwidth $E(\pi) - E(0)$ becomes narrower, signaling an increasingly heavier polaron; the quasiparticle weight decreases considerably, even at $k = 0$. Note that for this value of Ω_0 , even $\lambda = 2$ is still in the intermediary regime, with a ground-state quasiparticle weight $Z(0) \approx 0.32$.

We can now gauge the effects of increasing $\Delta\Omega$. Starting first with $E(k \sim 0)$, we see that this results in an increase in the polaron energy (for the same value of λ) and a significant additional flattening of the band, implying an even heavier effective mass (see following). Quantitatively, both these effects increase with increasing λ .

Near the edge of the Brillouin zone, we see a downturn of the dispersion relation which is more pronounced for smaller λ . This downturn is due to the fact that the polaron band must lie below the polaron + one-phonon continuum, which comprises excited states where the polaron scatters on one or

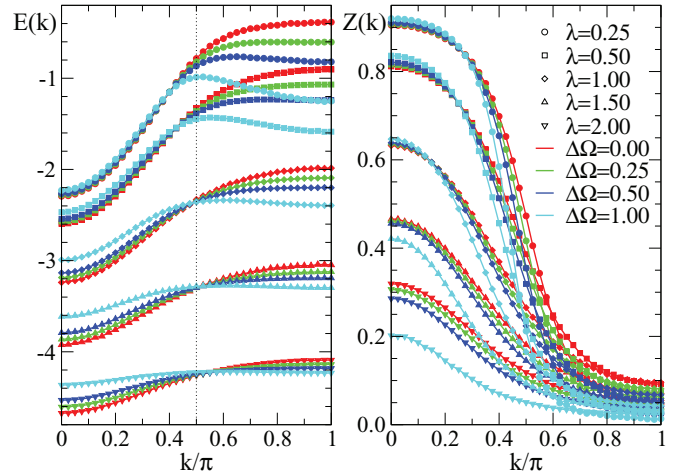


FIG. 1. (Color online) Polaron energy $E(k)$ and quasiparticle weight $Z(k)$ for $t = 1, \Omega_0 = 2$ and various phonon bandwidths (red, green, blue, and cyan are for $\Delta\Omega = 0.0, 0.25, 0.50$, and 1.00 , respectively) and effective couplings λ (with circles, squares, diamond, up triangles, and down triangles for $\lambda = 0.25, 0.50, 1.00, 1.50$, and 2.00 , respectively).

more phonons that do not belong to its cloud. The lower edge of this continuum is at $\min_q [E(k - q) + \Omega(q)]$. If $\Delta\Omega = 0$, this continuum starts at $E(0) + \Omega_0$ for all k . If $\Delta\Omega \neq 0$, its boundary varies with k . In particular, for $k = \pi$ and normal (negative) phonon dispersion, the minimum is reached at $E(0) + \Omega(\pi)$ where both terms are minimized. This agrees with the results of Fig. 1 and explains the apparent “folding” of the polaron dispersion. One immediate conclusion is that for dispersive phonons, one can not use the location of the continuum to fully determine the phonon energy.

The effect of increasing $\Delta\Omega$ on the $Z(k)$ is different in the weak- and strong-coupling regimes, for $k = 0$. Here, we see an increase of $Z(k \approx 0)$ with $\Delta\Omega$ at small coupling, while for stronger coupling λ it decreases with increasing $\Delta\Omega$. As we show below, the effective mass increases with increasing $\Delta\Omega$ for all λ . This warns against inappropriate use of the familiar relationship $m^*/m_0 = 1/Z(0)$, which only holds if the self-energy is momentum independent.¹ For the usual Holstein model with $\Delta\Omega = 0$, the momentum dependence of the self-energy is so weak that this equality is satisfied with reasonable accuracy. For dispersive phonons, however, the self-energy acquires significant momentum dependence [the downturn in $E(k \approx \pi)$ is another indication that this must be the case]. As a result, $m^*/m_0 = 1/Z(0)$ is no longer valid.

Figure 2(a) plots m^* versus λ for several values of $\Delta\Omega$. Symbols show m^* extracted from $E(k)$ by fitting it to $\sum_{n \leq 6} a_n \cos(nk)$. We find that m^* increases with increasing $\Delta\Omega$ for all λ , although for small λ the change is very small. To confirm this, we use perturbation theory.

As is usually the case for polaron problems, we find that the Rayleigh-Schrödinger approach yields better agreement than Wigner-Brillouin perturbation theory; we restrict ourselves to presenting results only from the former. For $\lambda \rightarrow 0$, we start with the free electron in a phonon vacuum state $|k\rangle$ and take the e-ph term as the weak perturbation. The first correction to

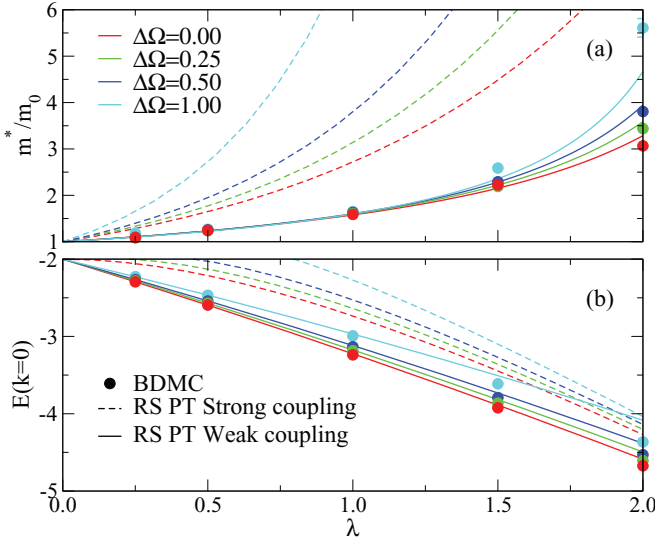


FIG. 2. (Color online) (a) Effective mass m^*/m_0 , and (b) polaron ground-state energy $E(0)$ vs the effective coupling λ for various phonon bandwidths $\Delta\Omega$. Symbols show BDMC results, with error bars smaller than the symbol size except where explicitly shown. Full lines are results from Rayleigh-Schrödinger second-order (i.e., fourth order in the electron-phonon coupling) perturbation theory for weak coupling, while dashed lines are results from Rayleigh-Schrödinger first-order perturbation theory for strong coupling.

the energy is

$$E^{(2)}(k) = \frac{1}{N} \sum_q \frac{g^2}{\epsilon(k) - \epsilon(k-q) - \Omega(q)}, \quad (4)$$

which for the dispersion of Eq. (2) gives

$$E^{(2)}(k) = \frac{-2t\lambda\Omega_0\sqrt{1-\delta}}{\sqrt{4t\Omega_0 \cos k + \Omega_0^2(1-\delta) - 4t^2 \sin^2 k}}. \quad (5)$$

From this, we find

$$\frac{m^*}{m_0} = \left[1 - \frac{\lambda t \Omega_0 (2\Omega_0 + 4t) \sqrt{1-\delta}}{[4t\Omega_0 + (1-\delta)\Omega_0^2]^{3/2}} \right]^{-1}. \quad (6)$$

The next correction $E^{(4)}(k)$ is calculated similarly (we do not write its long expression here). Comparison between BDMC (symbols) and perturbational results $E_{\lambda \rightarrow 0}(k) = \epsilon(k) + E^{(2)}(k) + E^{(4)}(k)$ (full lines) is shown in Fig. 2(b). The agreement is good up to $\lambda \sim 1$ and then becomes progressively worse. We note that although the perturbational expression works very well at $k \approx 0$ and small λ , it should not be trusted at larger k because the denominator of Eq. (4) vanishes at a finite k , resulting in unphysical behavior. For $k \rightarrow 0$, however, we can use it to calculate the perturbational prediction for m^* , shown by full lines in Fig. 2(a). It confirms that m^* increases with $\Delta\Omega$ although for small λ the effect is tiny. It is also straightforward to find $Z(k) = 1 - \alpha_k + \dots$ and the average number of phonons $N_{ph}(k) = \alpha_k + \dots$, where $\alpha_k = g^2/N \sum_q [\epsilon(k) - \epsilon(k-q) - \Omega(q)]^{-2}$.

For $\lambda \gg 1$, we take the electron kinetic energy as the small perturbation.¹⁴ For $t = 0$, the ground state is found using the

unitary transformation e^S where

$$S = - \sum_q \frac{g}{\Omega_q} [b_{-q}^\dagger - b_q] \frac{1}{\sqrt{N}} \sum_k c_{k+q}^\dagger c_k. \quad (7)$$

For $\Delta\Omega = 0$, this is the Lang-Firsov transformation.¹⁵ If the electron is located at site i , the transformed phonon annihilation operator is found to be

$$B_{q,i} = e^S b_q e^{-S} |_{\hat{n}_i=1} = b_q + \frac{g}{\Omega(q)} \frac{e^{-iqR_i}}{\sqrt{N}}. \quad (8)$$

Within this Hilbert subspace the Hamiltonian becomes

$$\hat{\mathcal{H}}_{t=0} |_{\hat{n}_i=1} = \sum_q \Omega(q) B_{q,i}^\dagger B_{q,i} - \frac{g^2}{\sqrt{\Omega_0(\Omega_0 - \Delta\Omega)}}. \quad (9)$$

The second term gives the ground-state energy in this limit. Its wave function is $|\tilde{i}\rangle = c_i^\dagger \exp[-\frac{g^2}{2N} \sum_q [\Omega(q)]^{-2} - \frac{g}{\sqrt{N}} \sum_q e^{-iqR_i} [\Omega(q)]^{-1} b_q^\dagger] |0\rangle$. Hopping lifts the degeneracy between these states and, to first order, leads to eigenstates $|\tilde{k}\rangle = \frac{1}{\sqrt{N}} \sum_i e^{ikR_i} |\tilde{i}\rangle$ of energy $E_{\lambda \rightarrow \infty}(k) = -2t\lambda - 2t^* \cos(k) + \dots$, where the effective hopping t^* is exponentially suppressed so that

$$\frac{m^*}{m_0} = \frac{t}{t^*} = e^{\frac{2\lambda}{\Omega_0(1-\delta)}} + \dots, \quad (10)$$

while $Z(k) \approx \exp[-N_{ph}]$ where the average number of phonons is $N_{ph} = [2 + \delta/(1-\delta)]t\lambda/\Omega_0 + \dots$.

The strong-coupling perturbational results are shown as dashed lines in Fig. 2. While trends are correct, even for $\lambda = 2$ the agreement is poor. This is not surprising because, as mentioned, $\lambda = 2$ is only an intermediate coupling for the adiabaticity ratio used here. Equation (10) confirms that m^* increases with $\Delta\Omega$ for a fixed λ . We also see that $m^*/m_0 \neq 1/Z(0)$ even when $\lambda \rightarrow \infty$.

These results suggest a reason for the larger polaron effective mass for (normally) dispersive optical phonons with $\delta = \Delta\Omega/\Omega_0 > 0$. The speculation that phonon mobility may lead to a more extended cloud and thus larger m^* is not borne out: phonons are mobile for any $\delta \neq 0$, yet BDMC results (not shown) and the perturbational formulas show that m^* decreases if $\delta < 0$. The difference is that if $\delta > 0$, the negative phonon dispersion leads to a phonon speed with a sign opposite to that of the carrier. For a polaron with small momentum $k > 0$, since the carrier must be close to its ground state $k_c \approx 0$, the contribution from the momenta of all the phonons in the cloud $\sum_i q_i = k - k_c$ must be small and positive. However, phonons with small positive momentum move in a direction opposite to that of the polaron, slowing it down (aside from being energetically costly). Less expensive phonons with positive speed have momenta just above $-\pi$, but balancing many such momenta to obtain a small positive total is challenging. If $\delta < 0$, however, phonons with small momentum move in the same direction as the carrier and are the least costly, so m^* decreases with $|\delta|$.

This explanation is also supported by the fact that m^* starts to change considerably only once the polaron bandwidth becomes smaller than the phonon bandwidth. If the phonons are much slower than the polaron, then the cloud will primarily move through phonon emission and absorption by the carrier,

like for $\delta = 0$, and m^* should not be much affected. Indeed, this is what we observe for small λ . This also suggests that, as far as m^* is concerned, $\Delta\Omega/t^*$ may be a more suitable dimensionless parameter to characterize the effects of phonon dispersion. However, this may not be true for all quantities so we continue to use δ as the third parameter.

For the usual Holstein model it is known that λ is the primary parameter that determines polaron properties, for example, the crossover to a small polaron occurs at $\lambda \sim 1$.⁸ The adiabaticity ratio influences the “sharpness” of the crossover and its precise location but does not lead to qualitative changes, at least not while Ω_0/t is away from the strongly adiabatic regime (which is not suitable for study with BDMC, in any event).

Our results suggest that the same conclusions apply for a finite phonon bandwidth δ if it is small enough to keep the phonons gapped (as is reasonable for optical phonons¹⁶). For gapped phonons, the polaron crossover is known to remain smooth even if $\delta \neq 0$,¹⁷ so δ may only affect its sharpness. We can estimate this by considering, for example, its influence on the evolution of $\ln m^*/m_0$ with λ . The slope of this quantity for both $\lambda \ll 1$ and $\lambda \gg 1$ can be obtained from perturbation theory. The larger the mismatch between the two, the sharper must be the crossover. If $\Omega_0/t \gg 1$, both Eqs. (6) and (10) give a slope of $2t/[\Omega_0(1 - \delta)]$, consistent with a very smooth crossover for both Einstein ($\delta = 0$) and gapped ($\delta < 1$) dispersive phonons. For $\Omega_0/t \ll 1$, the slope remains the same for $\lambda \gg 1$, while for $\lambda \ll 1$ we now find $\sqrt{t(1 - \delta)}/(4\Omega_0)$. The

mismatch is severe, explaining the expected sharp crossover in this limit for $\delta = 0$. If $0 < \delta < 1$, the mismatch is further accentuated, leading to an increasingly sharper crossover with increasing δ . Figure 2 shows the beginning of this crossover, especially when combined with the $\lambda \gg 1$ results (dashed lines).

Similar considerations allow us to speculate on the effects of δ for a dispersion similar to Eq. (2) in $D > 1$. Again, no significant changes with δ are expected if λ is small. For $\lambda \gg 1$, perturbation theory confirms an increase of m^* with δ and a sharpening of the crossover for $\Omega_0/t \ll 1$, even if these effects are slightly weaker than in 1D. For detailed quantitative investigations in 2D and 3D, it is probably best to use a realistic phonon dispersion.

To conclude, we studied the effect of dispersive optical phonons on the properties of a Holstein polaron. For weak e-ph coupling, m^* is little affected so the Einstein mode approximation is valid, although it may miss important higher-energy physics such as the downturn in $E(k)$ at the Brillouin zone edge. For larger e-ph coupling, however, m^* may increase (decrease) significantly if $\delta > 0$ ($\delta < 0$). This suggests that quantitative modeling of materials with strong e-ph coupling needs to explicitly take the dispersion of the optical phonons into account.

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