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Holstein polaron: The effect of coupling to multiple-phonon modes

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Abstract – We investigate the effects of coupling to multiple-phonon modes on the properties of a Holstein polaron. To this end, we generalize the Momentum Average approximations $MA^{(0)}$ and $MA^{(1)}$ to deal with multiple-phonon modes. As for a single-phonon mode, these approximations are found to be numerically very efficient. They become exact for very weak or very strong couplings, and are highly accurate in the intermediate regimes, *e.g.* the spectral weights obey exactly the first six, respectively eight, sum rules. Our results show that the effect on ground-state properties is cumulative in nature. As a result, if the effective coupling to one mode is much larger than to all the others, this mode effectively determines the ground-state properties. However, even very weak coupling to a second phonon mode has important non-perturbational effects on the higher-energy spectrum, in particular on the dispersion and the phonon statistics of the polaron band. This has important consequences on the analysis and interpretation of data for real materials.

The coupling of electrons to phonons is a widely studied problem, because it leads to many interesting phenomena such as conventional superconductivity or the formation of polarons (composite objects comprised of an electron and the surrounding phonon cloud), important in several classes of materials. As a recent example, results from angle-resolved photoemission spectroscopy [1] (ARPES) have lead to new discussions about possible polaronic effects in high-temperature superconductors [2].

Even though complex materials have many optical and acoustic phonons, most theoretical studies of polaron properties are of the Holstein model based on coupling to a single optical phonon mode [3,4]. One reason is that usually there is one optical mode to which the coupling is strongest, and one assumes that the effects of the other modes are perturbationally small. Equally importantly, the efficiency of various numerical methods [4] used to study the single-mode Holstein model in the intermediate coupling regime, where no exact solutions are known, suffers when the Hilbert space is enlarged by addition of multiple phonon modes. In fact, generalizations of numerical methods, such as path-integral Monte Carlo, that may efficiently deal with multiple phonon modes, are just being proposed [5]. To the best of our knowledge, there are no results available in the current literature discussing the effects on polaron properties coming from coupling to multiple-phonon modes.

Recently, the so-called Momentum Average (MA) analytical approximation [6,7] has been shown to be highly accurate over most of the parameter space of the single-mode Holstein polaron, while requiring a numerically trivial effort irrespective of the dimensionality of the problem or the strength of the coupling. Moreover, its accuracy can be systematically improved [8]. Of course, for an extensively studied problem such as the single-mode Holstein polaron, the MA just reproduced already known results. The hope is that this numerically efficient yet accurate approach can be extended to Hamiltonians for which there are few or no available numerical results. In such cases, MA would be useful for a quick yet accurate survey of properties in various regimes, which can then be followed by quantitatively more accurate, but significantly more time- and resource-consuming numerical simulations.

In this letter we demonstrate that MA can be generalized to deal with a Holstein polaron coupled to multiple-phonon modes, without loss of accuracy when compared to the single-mode results. This allows us to study, for the first time, the effects of coupling to multiple-phonon modes on the polaron properties, both ground-state and higher energy. Our results are the first to highlight the

importance of considering the coupling to multiple phonon modes in real materials. As we show below, while for ground-state properties these effects are rather trivial, the higher-energy spectrum is significantly modified by additional phonon modes, even if coupling to them is perturbationally small. This shows that neglect of all but the most strongly coupled phonon mode is thoroughly unjustified.

Starting from the second quantization form of the original Holstein Hamiltonian [9,10], we generalize it by considering multiple dispersionless phonon modes, Ω_α, which have local interactions with the electrons:

$$\mathcal{H} = \sum_{\mathbf{k}} \epsilon_{\mathbf{k}} c^\dagger_{\mathbf{k}} c_{\mathbf{k}} + \sum_{\mathbf{q},\alpha} \Omega_\alpha b^{\alpha\dagger}_{\mathbf{q}} b^\alpha_{\mathbf{q}} + \sum_{\alpha,\mathbf{k},\mathbf{q}} \frac{g_\alpha}{\sqrt{N}} c^\dagger_{\mathbf{k}-\mathbf{q}} c_{\mathbf{k}} (b^{\alpha\dagger}_{\mathbf{q}} + b^\alpha_{-\mathbf{q}}). \quad (1)$$

The first term describes a free electron on a d-dimensional lattice with N sites and periodic boundary condition. The spin of the electron is irrelevant, thus the spin index is dropped. We use a free-electron dispersion corresponding to nearest-neighbor hopping on a d-dimensional, simple cubic-like lattice of constant $a=1$, though this can be trivially generalized to any other dispersion. The second term describes the optical phonon modes with energies Ω_α, and the third term describes Holstein-like electron-phonons couplings, characterized by coupling constants g_α. Momenta sums are over the Brillouin zone.

We focus on finding the polaron's Green's function [6,7]:

$$G(\mathbf{k},\omega) = \langle 0 | c_{\mathbf{k}} \hat{G}(\omega) c^\dagger_{\mathbf{k}} | 0 \rangle, \quad (2)$$

where $\hat{G}(\omega) = [\omega - \mathcal{H} + i\eta]^{-1}$ is the usual resolvent ($\hbar = 1$) and $|0\rangle$ is the vacuum. The poles of this Green's function mark the polaron spectrum, and ground-state (GS) energies, effective masses, quasiparticle (qp) weights and average phonon numbers can then be calculated as discussed in ref. [7]. The spectral weight $A(\mathbf{k},\omega) = -\frac{1}{\pi} G(\mathbf{k},\omega)$ can also be directly compared against ARPES results.

To simplify notation, we first assume that there are only two phonon modes, and rename their operators as $b_{\mathbf{q}}$ (mode 1) and $B_{\mathbf{Q}}$ (mode 2). The generalization to more phonon modes is discussed below. We use repeatedly Dyson's identity $\hat{G}(\omega) = \hat{G}_0(\omega) + \hat{G}(\omega)\hat{V}\hat{G}_0(\omega)$, where $\hat{G}_0(\omega) = [\omega - \mathcal{H}_0 + i\eta]^{-1}$ corresponds to the non-interacting Hamiltonian, to generate an infinite system of coupled equations involving $G(\mathbf{k},\omega)$ and the generalized Green's functions:

$$F_{nm}(\mathbf{k},\mathbf{q}_1,\ldots,\mathbf{q}_n;\mathbf{Q}_1,\ldots,\mathbf{Q}_m;\omega) = \langle 0 | c_{\mathbf{k}} \hat{G}(\omega) c^\dagger_{\mathbf{k}_T} b^\dagger_{\mathbf{q}_1} \ldots b^\dagger_{\mathbf{q}_n} B^\dagger_{\mathbf{Q}_1} \ldots B^\dagger_{\mathbf{Q}_m} | 0 \rangle.$$

Here $\mathbf{k}_T = \mathbf{k} - \mathbf{q}_T - \mathbf{Q}_T$, where $\mathbf{q}_T = \sum_{i=1}^n \mathbf{q}_i$ and $\mathbf{Q}_T = \sum_{j=1}^m \mathbf{Q}_j$ are the total momenta carried by the two types of phonons. Arguments identical to those of ref. [8] show that all functions F_{nm} are proportional to $G(\mathbf{k},\omega)$, because the only inhomogeneous term in their recurrence relations is proportional to this quantity. It is thus more convenient to work with the rescaled functions:

$$f_{nm}(\mathbf{k},\{\mathbf{q}\},\{\mathbf{Q}\},\omega) = N^{\frac{n+m}{2}} \frac{F_{nm}(\mathbf{k},\{\mathbf{q}\},\{\mathbf{Q}\},\omega)}{G(\mathbf{k},\omega)},$$

where we introduce the shorthand notations $\{\mathbf{q}\} \equiv \mathbf{q}_1,\ldots,\mathbf{q}_n$, $\{\mathbf{q}\}_i \equiv \mathbf{q}_1,\ldots,\mathbf{q}_{i-1},\mathbf{q}_{i+1},\ldots,\mathbf{q}_n$ and $\{\mathbf{q}\}_{n+1} \equiv \mathbf{q}_1,\ldots,\mathbf{q}_n,\mathbf{q}_{n+1}$. The functions f_{nm} are then given by the recurrence relations

$$\begin{aligned} f_{nm}(\{\mathbf{q}\},\{\mathbf{Q}\}) &= G_0(\mathbf{k}_T, \omega - n\Omega_1 - m\Omega_2) \\ &\times \Bigg[g_1 \sum_{i=1}^n f_{n-1,m}(\{\mathbf{q}\}_i,\{\mathbf{Q}\}) + g_2 \sum_{j=1}^m f_{n,m-1}(\{\mathbf{q}\},\{\mathbf{Q}\}_j) \\ &+ \frac{g_1}{N} \sum_{\mathbf{q}_{n+1}} f_{n+1,m}(\{\mathbf{q}\}_{n+1},\{\mathbf{Q}\}) \\ &+ \frac{g_2}{N} \sum_{\mathbf{Q}_{m+1}} f_{n,m+1}(\{\mathbf{q}\},\{\mathbf{Q}\}_{m+1}) \Bigg], \end{aligned} \quad (3)$$

where $f_{00} \equiv 1$ by definition, the dependence on $\mathbf{k},\omega$ is implicitly assumed for all other f_{nm}, and $G_0(\mathbf{k},\omega) = (\omega - \epsilon_{\mathbf{k}} + i\eta)^{-1}$ is the free propagator.

The Green's function has the standard form

$$G(\mathbf{k},\omega) = [\omega - \epsilon_{\mathbf{k}} - \Sigma(\mathbf{k},\omega) + i\eta]^{-1}, \quad (4)$$

where the exact self-energy is given by

$$\Sigma(\mathbf{k},\omega) = \frac{g_1}{N} \sum_{\mathbf{q}_1} f_{10}(\mathbf{k},\mathbf{q}_1,\omega) + \frac{g_2}{N} \sum_{\mathbf{Q}_1} f_{01}(\mathbf{k},\mathbf{Q}_1,\omega), \quad (5)$$

and $f_{10}(\mathbf{k},\mathbf{q}_1,\omega)$ and $f_{01}(\mathbf{k},\mathbf{Q}_1,\omega)$ are the solutions of the above set of recurrence relations, eqs. (3).

As for the single-mode problem, the MA$^{(0)}$ approximation is obtained by replacing the free propagator with its momentum average over the Brillouin zone, in the r.h.s. of each of eqs. (3):

$$G_0(\mathbf{k}_T, \omega - n\Omega_1 - m\Omega_2) \to \bar{g}_0(\omega - n\Omega_1 - m\Omega_2) \equiv \bar{g}_0(\omega_{nm}),$$

where from now we use the shorthand notation

$$\omega_{nm} = \omega - n\Omega_1 - m\Omega_2, \quad (6)$$

and the momenta averages over the Brillouin zone

$$\bar{g}_0(\omega) = \frac{1}{N} \sum_{\mathbf{k}} G_0(\mathbf{k},\omega)$$

are simple known functions[1]. The resulting simplified recurrence relations can be solved in terms of the momentum averaged functions

$$\mathcal{F}_{nm}(\omega) = \frac{1}{N^{m+n}} \sum_{\{\mathbf{q}\},\{\mathbf{Q}\}} f_{nm}(\mathbf{k},\{\mathbf{q}\},\{\mathbf{Q}\},\omega). \quad (7)$$

[1] Expressions of $\bar{g}_0(\omega)$ for nearest-neighbor hopping on simple cubic lattice in $d = 1,2,3$ are listed in ref. [7].

Thus, the momentum-independent $\mathrm{MA}^{(0)}$ self-energy is

$$\Sigma_{MA^{(0)}}(\omega) = g_1\mathcal{F}_{10}(\omega) + g_2\mathcal{F}_{01}(\omega),$$

while the recurrence relations (3) become

$$\begin{aligned}\mathcal{F}_{nm}(\omega) \;=\; & \bar{g}_0(\omega_{nm})[ng_1\mathcal{F}_{n-1,m}(\omega) + mg_2\mathcal{F}_{n,m-1}(\omega) \\ & + g_1\mathcal{F}_{n+1,m}(\omega) + g_2\mathcal{F}_{n,m+1}(\omega)]. \end{aligned} \tag{8}$$

Of course, $\mathcal{F}_{00} = 1$.

Similar recursive equations were previously solved in a different context by Cini *et al.* [11,12]. They rewrite these as matrix equations, in terms of vectors $V_n = (\mathcal{F}_{n,0}, \mathcal{F}_{n,1}, \ldots)^T$, *i.e.* vectors containing all unknowns with a fixed number of phonons of type 1. The solution is then a continued fraction of matrices of dimension equal to the maximum allowed number of second-type phonons. However, their solution cannot be generalized to more than two phonon modes, nor to the equations resulting for $\mathrm{MA}^{(1)}$ or higher levels of the approximation (see below). We have found an alternative solution without these shortcomings. First, we rewrite these recurrence relations in matrix form:

$$V_n = A_nV_{n-1} + B_nV_{n+1}, \tag{9}$$

where the vector $V_n = (\mathcal{F}_{n,0}; \mathcal{F}_{n-1,1}; \ldots; \mathcal{F}_{1,n-1}; \mathcal{F}_{0,n})^T$ contains all $n+1$ functions corresponding to a total of n phonons. A_n is a matrix of size $n+1 \times n$ with the only non-zero elements $(A_n)_{i,i} = (n-i)g_1\bar{g}_0(\omega_{n-i,i})$ and $(A_n)_{i+1,i} = (i+1)g_2\bar{g}_0(\omega_{n-1-i,i+1})$, $\forall i = 0, n-1$. Similarly, B_n is a matrix of size $n+1 \times n+2$ with the only finite elements $(B_n)_{i,i} = g_1\bar{g}_0(\omega_{n-i,i})$ and $(B_n)_{i,i+1} = g_2\bar{g}_0(\omega_{n-i,i})$, $\forall i = 0, n$. Dependence on ω is again implicitly assumed everywhere.

The solution is $V_n = M_nV_{n-1}$ with $V_0 = (1)$, where

$$M_n = \cfrac{1}{1 - B_n \cfrac{1}{1 - B_{n+1}\cfrac{1}{1-\ldots}A_{n+2}}A_{n+1}}A_n, \tag{10}$$

is a continued fraction of matrices of increasing size. The self-energy is $\Sigma_{MA^{(0)}}(\omega) = (g_1, g_2)V_1 = (g_1, g_2)M_1$. The continued fractions become convergent if truncated at levels $N > \max(g_1^2/\Omega_1^2, g_2^2/\Omega_2^2)$, *i.e.* when one keeps contributions from f_{nm} corresponding to expected average numbers of phonons in the cloud. The generalization to more phonon modes is straightforward. V_n again contains all Green's functions with a fixed number of phonons, and the interaction links it only to $V_{n\pm1}$. The matrices A_n and B_n have non-vanishing elements only on a number of diagonals equal to the number of modes. The dimension of these matrices increases now faster with increasing n, but this is still much less severe than the corresponding increase in numerical simulations. Also, note that the MA calculation is equally simple in any dimension, the only change being in the expression used for $\bar{g}_0(\omega)$ (see footnote [1]).

The analysis of the diagrammatic and variational meaning of $\mathrm{MA}^{(0)}$ and the sum rules it obeys, is identical to that for the single-mode case [6–8], and we do not repeat it. It again proves its accuracy over the entire parameter space, as long as $\Omega_i/t > 0.1, \forall i$. However, $\mathrm{MA}^{(0)}$ fails to correctly predict the polaron + one phonon continuum [7,13,14]. To remedy this, we need to use $\mathrm{MA}^{(1)}$ or a higher level approximation [8]. In $\mathrm{MA}^{(1)}$, eqs. (3) for f_{01} and f_{10} are left unchanged (*i.e.* exact), and the momentum average is made only for f_{nm} with $n+m \geqslant 2$. As shown in ref. [8], $\mathrm{MA}^{(1)}$ correctly predicts the polaron + one phonon continuum, besides giving small improvements in the accuracy of various other quantities, *e.g.* the number of exactly satisfied sum rules increases from six to eight.

The derivation of $\Sigma_{MA^{(1)}}(\omega)$ for the multiple-mode case is similar to that in ref. [8], with the only difference that continued fractions there correspond to continued fractions of matrices (like in eq. (10)) here. We again introduced the total momentum averages quantities $\mathcal{F}_{nm}$, as in eq. (7), but we also need to introduce two partially momentum-averaged quantities, namely for $n \geqslant 1$

$$\delta\overline{\mathcal{F}_{nm}}(\mathbf{q}_1) = \frac{1}{N^{n-1+m}} \sum_{\{\mathbf{q}\}_1, \{\mathbf{Q}\}} f_{nm}(\{\mathbf{q}\}, \{\mathbf{Q}\}) - \mathcal{F}_{nm}$$

and for $m \geqslant 1$

$$\delta\underline{\mathcal{F}_{nm}}(\mathbf{Q}_1) = \frac{1}{N^{n-1+m}} \sum_{\{\mathbf{q}\}, \{\mathbf{Q}\}_1} f_{nm}(\{\mathbf{q}\}, \{\mathbf{Q}\}) - \mathcal{F}_{nm}$$

(dependence on $\mathbf{k}, \omega$ is again implicitly assumed). The equations of recurrence for $\mathcal{F}_{nm}$, $n+m \geqslant 2$, are the same and therefore solved as before. It is straightforward to verify that $\delta\overline{\mathcal{F}_{nm}}(\mathbf{q}_1)$ and $\delta\underline{\mathcal{F}_{nm}}(\mathbf{Q}_1)$ satisfy recurrence relations very similar to those of $\mathcal{F}_{nm}$. In fact, the matrix B_n stays the same in both cases, but the corresponding matrices A_n are changed by decreasing by 1 the number of phonons of type 1 or 2, respectively. Thus, the solutions for $\delta\overline{\mathcal{F}_{nm}}$, $\delta\underline{\mathcal{F}_{nm}}$ involve continued fractions of matrices $\overline{M_n}$ and $\underline{M_n}$ similar to eq. (10) but with the appropriate A_i. After some algebraic manipulations, the $\mathrm{MA}^{(1)}$ self-energy can be written as

$$\begin{aligned}\Sigma_{MA(1)}(\omega) &= \begin{pmatrix} g_1 & g_2 \end{pmatrix} \\ &\times \left[1 - \begin{pmatrix} g_1\gamma_1 & g_2\gamma_2 & 0 \\ 0 & g_1\gamma_2 & g_2\gamma_1 \end{pmatrix} M_2\right]^{-1} \begin{pmatrix} g_1\gamma_1 \\ g_2\gamma_2 \end{pmatrix}, \end{aligned} \tag{11}$$

where $\gamma_i = \bar{g}_0(\tilde{\omega}_i)/[1 + (g_1a_1^i + g_2a_2^i)\bar{g}_0(\tilde{\omega}_i)]$, the renormalized energies are $\tilde{\omega}_i = \omega - (g_1a_1^i + g_2a_2^i) - \Omega_i$, and the coupling constant factors are $a_1^1 = (\overline{M_2})_{0,0}$, $a_2^1 = (\overline{M_2})_{0,1}$, $a_1^2 = (\underline{M_2})_{1,1}$ and $a_2^2 = (\underline{M_2})_{2,1}$. Like for the single-mode Holstein polaron, we find that $\Sigma_{MA^{(1)}}(\omega)$ is independent of $\mathbf{k}$; momentum dependence only arises from $\mathrm{MA}^{(2)}$ or higher levels. More details on this derivation and the efficient generalization to three or more phonon modes will be presented elsewhere [15].

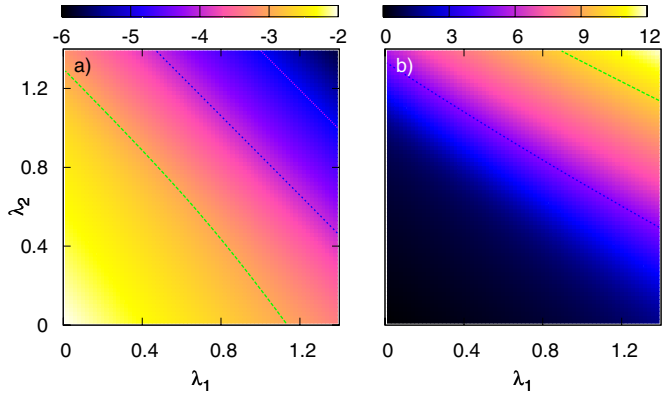

Fig. 1: (Color online) (a) $\mathrm{MA}^{(1)}$ GS energy and (b) $\ln(m^*/m)$, where m^* is the polaron effective mass, *vs.* coupling constants λ_1 and λ_2, for $\Omega_1 = 0.7t$, $\Omega_2 = 0.3t$ and $d = 1$.

All results shown below are for two phonon modes and $d = 1$, which suffices to uncover the essential new physics. Results for higher d and more phonon modes are qualitatively similar and will be presented elsewhere [15]. We begin by discussing GS properties such as the energy E_{GS} and effective mass m^*, shown in fig. 1 as functions of the effective couplings $\lambda_i = g_i^2/(2dt\Omega_i)$, $i = 1, 2$. Note that the *qp* weight is related to the effective mass: $Z_0 = m/m^*$, where m is the bare electron mass, because the $\mathrm{MA}^{(1)}$ self-energy is momentum independent [7]. The "equipotential" lines drawn show that E_{GS} is well described as a function of only $\lambda_{\text{eff}} = \sum_i \lambda_i$, whereas m_* is a function of $\sum_i \lambda_i/\Omega_i = 2dt \sum_i g_i^2/\Omega_i^2$. If $\Omega_i = \Omega$ for all modes, these can be proved to be *exact* results [15]. In the strong-coupling limit one also expects $E_{GS} = -\sum_i g_i^2/\Omega_i$, $\ln Z_0 \propto -\sum_i g_i^2/\Omega_i^2$, supporting the same conclusion. To a good extent the only effect of having $\Omega_1 \neq \Omega_2$ is to change the slope of the m^* "equipotentials" from the 45° found when $\Omega_1 = \Omega_2$, although some slight deviations from linearity are also seen in E_{GS} when either $\lambda_i \ll 1$.

We conclude that GS properties can be well understood in cumulative terms, for instance the energy is well approximated by that of a polaron coupled to a single phonon with λ_{eff}. The crossover from large to small-polaron behavior is therefore expected when $\lambda_{\text{eff}} \approx 1$ [3,4]. As a result, it is possible to have small-polaron behavior even if each individual phonon mode is weakly coupled to the electron (each $\lambda_i < 1$, but $\sum_i \lambda_i > 1$). However, in cases where one mode (say, mode 1) is indeed much more strongly coupled than all others, $\lambda_1 \gg \lambda_i$, $i = 2, \ldots$, then $\lambda_{eff} \approx \lambda_1$ and one can, to a good extent, ignore the small cumulative effect from the other modes.

This conclusion, however, does not generally hold for higher-energy properties, as we show now. In fig. 2 we plot the $d = 1$ spectral function $A(k, \omega)$ *vs.* k and ω. The effective coupling to the first mode, of frequency $\Omega_1/t = 0.7$, is kept constant to a fairly low value $\lambda_1 = 0.4$. In panel (a), coupling to the second mode, of energy $\Omega_2/t = 0.3$, is zero, so this is in fact a one-phonon problem. As expected,

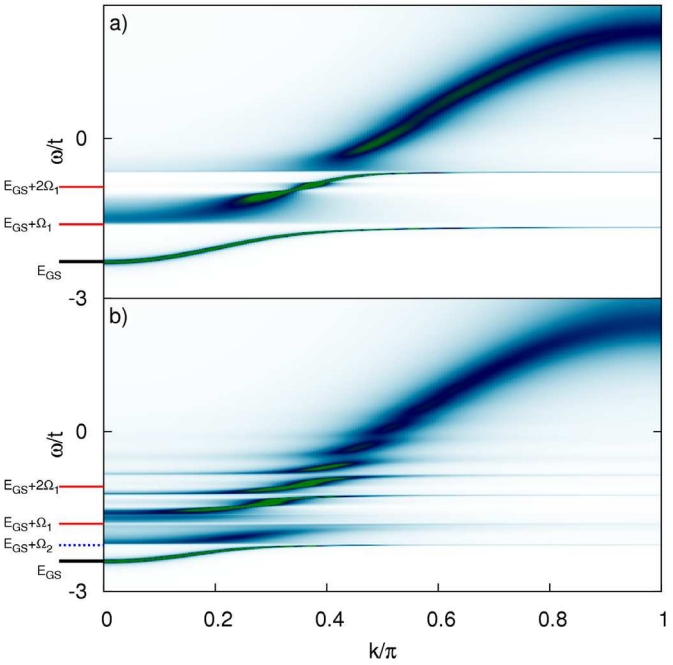

Fig. 2: (Color online) $A(k, \omega)$ from $\mathrm{MA}^{(1)}$ for $d = 1$, and for $\Omega_1 = 0.7t$, $\Omega_2 = 0.3t$, $\lambda_1 = 0.4$ and a) $\lambda_2 = 0.0$, b) $\lambda_2 = 0.2$.

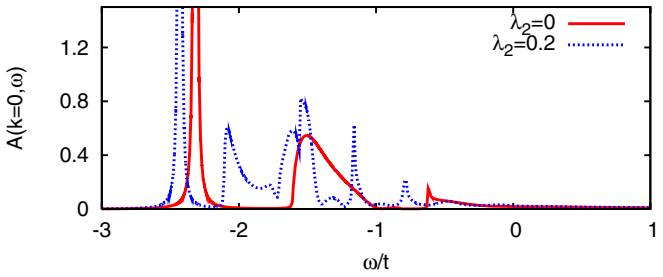

Fig. 3: (Color online) $A(k = 0, \omega)$ from $\mathrm{MA}^{(1)}$ for $d = 1$, and for $\Omega_1 = 0.7t$, $\Omega_2 = 0.3t$, $\lambda_1 = 0.4$.

at low energies we see the polaron band, of width Ω_1, followed above $E_{GS} + \Omega_1$ by the polaron + one-phonon continuum, and more features at higher energies [3,4,8].

Addition of even very weak coupling to a second phonon mode changes things considerably, as shown in panel (b) for $\lambda_2 = 0.2$. If $\Omega_2 < \Omega_1$ (as chosen here), the polaron band width is changed to Ω_2, even though the GS energy and effective mass are not much affected (see previous discussion). To see this effect more clearly we plot in fig. 3 the full spectral function $A(k = 0, \omega)$, while keeping λ_1 constant. Clearly, the first feature above the GS appears at Ω_2, not Ω_1, as soon as $\lambda_2 > 0$. This significant change is not so surprising if one considers the origin of the polaron + one phonon continuum: it corresponds to states where one phonon is created far from the polaron. As a result, they interact little and the total energy is just the sum of the two. If there are several phonon modes, the continuum will be defined by the mode with the lowest frequency Ω_{min}, irrespective of whether this is the mode

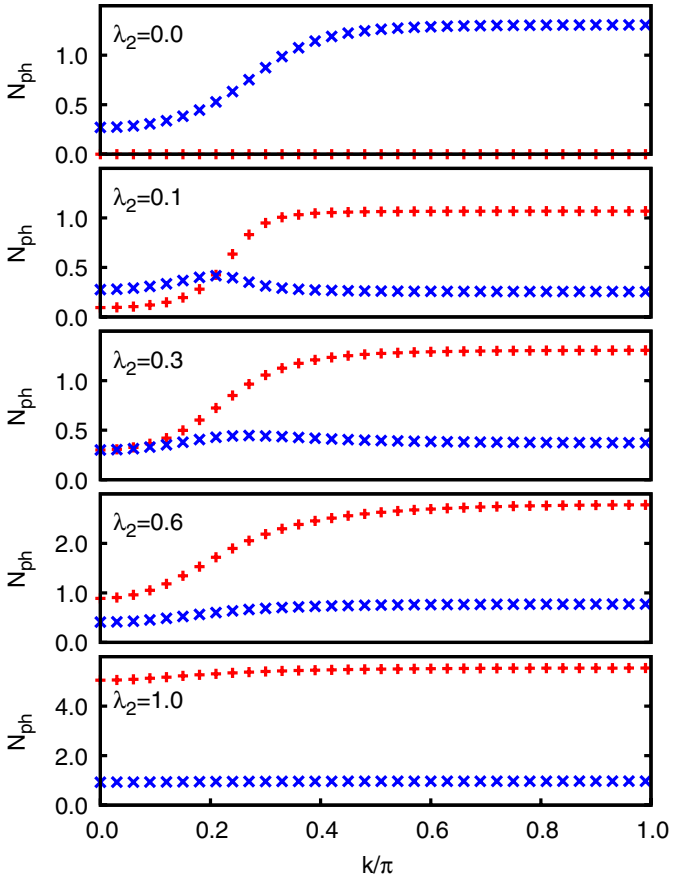

Fig. 4: (Color online) Number of phonons of both type 1 (× symbols) and type 2 (+ symbols) in the polaron cloud *vs.* λ_2, for $\Omega_1 = 0.7t$, $\Omega_2 = 0.3t$ and $\lambda_1 = 0.4$.

most strongly coupled to the electron or not. Because of this, the polaron band cannot be wider than $\Omega_{\rm min}$. While this argument is very reasonable, it seems to have been completely missed in literature discussions related to effects of phonons on electron spectra (see below).

This interpretation is confirmed by phonon statistics, shown in fig. 4. Here we plot average numbers N_1 and N_2 of phonons of either type in the polaron cloud, as a function of the polaron momentum k. These are calculated using the Hellman-Feynman theorem, $N_i(k) = \partial E_k/\partial\Omega_i$ [7,16]. Again, the coupling to the first mode is kept constant at $\lambda_1 = 0.4$. If $\lambda_2 = 0$ (first panel), we see that N_1 increases from a small value at $k = 0$ to just above 1 for $k > \pi/2$. This shows, as expected, that while around $k = 0$ the large-polaron is essentially similar to a free electron, for $k > \pi/2$ the largest contribution to the polaron comes from electron + one phonon states. Of course, $N_2 = 0$ in this case. As λ_2 is turned on but is still small ($\lambda_2 = 0.1, 0.3$, second and third panels) there is little change near $k = 0$, however the changes at higher momenta are dramatic: N_1 decreases by 1 whereas N_2 increases by 1. This confirms that it is now the second type of phonon that controls the nature of the polaron at large k values, even though $\lambda_2 < \lambda_1$. Once $\lambda_2 > \lambda_1$, the second phonon

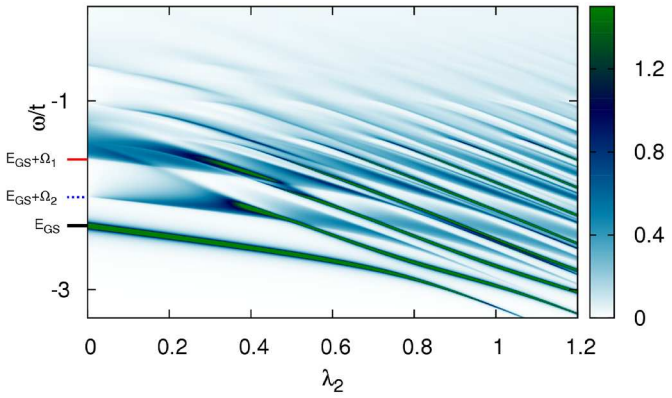

Fig. 5: (Color online) $A(k = 0, \omega)$ *vs.* ω and λ_2, for $\Omega_1 = 0.7t$, $\Omega_2 = 0.3t$ and $\lambda_1 = 0.4t$. For all $\lambda_2 > 0$, the continuum starts at $\Omega_2 < \Omega_1$.

completely dominates the behavior. For $\lambda_2 = 0.6$ (fourth panel) we have $\lambda_{\rm eff} = \lambda_1 + \lambda_2 = 1$, and the polaron is in the crossover regime, whereas for $\lambda_2 = 1.0, \lambda_{\rm eff} = 1.4$ the polaron is firmly in the small-polaron regime. In the latter case (fifth panel), we expect to see less and less k dependence, since the polaron cloud becomes limited to the site on which the electron resides. This is indeed observed for N_2, but also for N_1, because once the polaron is localized at a site (because of strong-coupling to mode 2) it will automatically also shift the equilibrium position for mode 1 phonons at that site, resulting in the creation of a finite number of such bare phonons.

We conclude that the polaron bandwidth at weak and intermediate couplings is given by the energy of the phonon of smallest frequency, irrespective of whether this is the most strongly coupled phonon or not. We now briefly discuss the consequences of this result.

If there is only one optical phonon mode coupled to the electron, one can use ARPES data to extract its frequency Ω from the location of the "discontinuity" in the dispersion, at weak coupling, or from average distance between eigenstates at strong couplings, where the Lang-Firsov spectrum appears [17]. Since the effective mass determines λ, g can then be extracted from a measurement of m^*. Our results show that this procedure is usually wrong in the case of multiple phonon modes, even if one expects coupling to one of them to be dominant. It only works if this particular mode also happens to have the lowest frequency, else one will underestimate its Ω and overestimate g. This point is clearly demonstrated in fig. 5, which shows that the $\Omega_{\rm min}$ phonon defines the location of the low-energy features above the GS, irrespective of its coupling. Of course, one may hope to see the continua due to the other phonon modes (see also fig. 2(b)) and thus be able to identify their frequencies. This is probably unlikely, due to broadening in real data (note that temperature dependence would also be determined by the $\Omega_{\rm min}$ phonon, not by the dominant one). Even if the Ω_i are identified from ARPES data, finding all g_i is generally impossible,

unless we know that one dominates, and we already know which one that is, *i.e.* we know its frequency from other considerations. The only simple case to interpret is the unlikely (for real materials) case where *all* phonons have roughly equal frequencies, when one can treat them as a single mode with coupling $g_{\text{eff}}^2 = \sum_i g_i^2$. In any event, it is clear that one cannot ignore the effects of coupling to multiple phonon modes, when trying to understand the properties of any real, complex materials where polaronic effects are thought to be important.

To summarize, we have found a generalization of the simple, yet accurate Momentum Average approximations to the problem of Holstein-type coupling to multiple phonon modes. This allows us to present the first ever accurate results for polarons coupled to multiple optical phonon modes, to the best of our knowledge. The possibility to obtain both low- and high-energy states gives a complete picture of the effects of the electron-phonon interactions in the presence of multiple phonon modes. Our results show that even perturbationally weak coupling to a second phonon can lead to essential changes of the spectral weight, if this phonon's frequency is less than that of the dominant phonon. In such cases, a simple-minded way to extract the electron-phonon coupling parameters from ARPES data is likely to lead to wrong values.

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