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Sharp transition for single polarons in the one-dimensional Su-Schrieffer-Heeger model

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We study a single polaron in the Su-Schrieffer-Heeger (SSH) model using four different techniques (three numerical and one analytical). Polarons show a smooth crossover from weak to strong coupling, as a function of the electron-phonon coupling strength $\lambda$, in all models where this coupling depends only on phonon momentum $q$. In the SSH model the coupling also depends on the electron momentum $k$; we find it has a sharp transition, at a critical coupling strength $\lambda_c$, between states with zero and nonzero momentum of the ground state. All other properties of the polaron are also singular at $\lambda = \lambda_c$. This result is representative of all polarons with coupling depending on $k$ and $q$, and will have important experimental consequences (eg., in ARPES and conductivity experiments).

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Polarons have been of broad interest in physics ever since they were introduced in 1933, to describe dielectric charge carriers \[1\]. Apart from their central role in solid-state physics, with many models now in use \[2,3\], they exemplify in quantum field theory the passage from weak to strong coupling in a non-trivial model of a single particle coupled to a bosonic field \[3\]. The first serious non-perturbative studies by Feynman \[5\] of the Frohlich polaron, are now a classic, but only recently were accurate results established across the whole range of coupling strengths \[6\]. Since then, exact numerical studies have been made of, eg., D-dimensional Holstein polarons in various lattice geometries, with $D = 1, 2, 3$ \[8\]; of 3D Rashba-Pekar polarons with short-range interactions \[8\]; of pseudo Jahn-Teller polarons \[11\]; and so on.

A central question in this field has been whether a sharp transition can exist in the polaronic ground state as a function of the dimensionless effective particle-boson coupling $\lambda$. In all the above-cited work there is simply a smooth crossover, expected when the coupling depends only on the bosonic momentum $q$; then there must always be non-zero matrix elements between the ground state and excited polaron states \[11\]. However, quite generally, one expects the coupling to depend on both $q$ and the particle momentum $k$; and then much less is known.

In this paper we study a specific example of this general case. The particle-boson coupling is taken from the well-known “SSH model”, introduced to describe electrons in 1-d polyacetylene \[12\]. Here we focus on the single polaron limit, not the more common case of half-filling, and the bosons are chosen to describe optical phonons. While this ignores the acoustic phonons which exist in real materials, it allows a direct comparison with the large number of results known for models which have a purely $q$-dependent coupling. The Hamiltonian thus takes the simple form $\mathcal{H} = H_o + V + H_{ph}$, where

$$H_o = -t_o \sum_i (c_i^\dagger c_{i+1} + h.c.) = \sum_k \epsilon_k c_k^\dagger c_k^\vphantom{\dagger},$$

describes the hopping of electrons between sites, with band dispersion $\epsilon_k = -2t_o \cos(k)$ ($c_i^\dagger$ creates an electron on site $i$; $c_k^\dagger$ creates a momentum state $k$). The term $H_{ph} = \omega_{ph} \sum_i b_i^\dagger b_i$ describes dispersionless phonons ($b_i^\dagger$ creates a phonon on site $i$). The interaction is

$$V = -\tilde{\alpha} t_o \sum_i (\hat{X}_i - \hat{X}_{i+1})(c_i^\dagger c_{i+1} + h.c.) = N^{-1/2} \sum_{k,q} M(k,q) c_{k+1}^\dagger c_k^\dagger (b_{-q}^\dagger + b_q^\vphantom{\dagger})$$

with site displacements $\hat{X}_i = \sqrt{\frac{\hbar}{2 M \omega_{ph}}} (b_i + b_i^\dagger)$, and the interaction vertex

$$M(k,q) = 2i\alpha [\sin(k+q) - \sin(k)] = i(2\omega_{ph} t_o)^{1/2} [\sin(k+q) - \sin(k)]$$

This interaction, with associated energy $\alpha = \tilde{\alpha} t_o \sqrt{\frac{\hbar}{2 M \omega_{ph}}}$, describes the modulation of the hopping amplitude by phonons. We henceforth set $t_o = 1$, and define two dimensionless parameters: the electron-phonon coupling parameter $\lambda = 2\alpha^2/(t_o \omega_{ph}) = \langle |M(k,q)|^2 \rangle/(2t_o \omega_{ph})$, where $\langle \cdot \rangle$ averages over the Brillouin zone; and the ‘adiabaticity’ ratio $\omega_{ph}/t_o = \omega_{ph}$ when $t_o = 1$.

(i) Results: We treat this non-perturbative problem with the Momentum Average (MA) analytical approximation \[6,13,14\] and three different numerical techniques: the Diagrammatic Monte Carlo (DMC) \[7\], the Limited Phonon Basis Exact Diagonalization (LPBED)
the curves in Fig. 1(e) look quite similar to those for $k$ where

$$k = \text{momentum}$$

and the renormalized effective polaron mass $m^*(\lambda) = (\partial^2 E(\lambda)/\partial k^2)^{-1}\big|_{k=k_G}$ (Fig. 2(d)), all show a sharp transition at $\lambda = \lambda_c(\omega_{ph})$ (see Fig. 2 for $\omega_{ph} = 0.5$, Fig. 3 for $\omega_{ph} = 3$). At this singularity, the polaronic mass $m^*(\lambda)$ diverges, with corresponding jumps in the first derivatives $dE_{GS}(\lambda)/d\lambda$ and $dZ_{GS}(\lambda)/d\lambda$, and in $d^2E_{GS}(\lambda)/d\lambda^2$. However the average number of phonons $N_{ph}(\lambda)$ in the polaronic polarization cloud does not diverge at $\lambda_c$ (although it is presumably still singular); $N_{ph}(\lambda_c) < 15$ for all values of the adiabaticity parameter $\omega_{ph}$ checked so far. Note also how $\lambda_c$ varies with $\omega_{ph}$ (Fig. 4), initially increasing for small $\omega_{ph}$, but then falling to the asymptotic value $\lambda_c \to 1/2$ in the instantaneous phonon limit $\omega_{ph} \to \infty$. This limit can be derived analytically (see below).

We emphasize here the remarkable agreement obtained between all 4 methods. The three numerical techniques are in principle exact, but all have their practical limitations, such as the sign problem noted below for QMC methods.

(ii) Discussion: The key new feature of couplings $M(k,q)$ like that in Eqs. (2) and (3), compared to $k$-independent couplings, is that they are non-diagonal in site index. Thus phonons cause the bandwidth to fluctuate, and can by themselves generate hopping between sites. The lowest-order process contributing to $E_{GS}$ is 2nd order in $M(k,q)$; higher corrections come from even powers of $M$. The same applies to the polaron self-energy, the polaron mass, quasiparticle renormalization, etc. Consider now a pair of vertices, connected by a
a 'sign problem' in any Monte Carlo calculation (indeed, refers to the instantaneous limit refer to MA and LPBED methods. The horizontal dotted line ω

FIG. 3: (Color online) The same as Fig. 2 but for ω_{ph} = 3. MA results in (b), (c) are shown as green solid lines.

FIG. 4: (Color online) Phase boundary dividing GSs with zero and nonzero momentum. Green squares and red triangles refer to MA and LPBED methods. The horizontal dotted line refers to the instantaneous limit \lambda_c^{\omega_{ph}\rightarrow \infty} = 1/2.

phonon of momentum \(q\); we have

\[ M_{k,-q}M_{k'-q, q} \propto \lambda \sin^2 \left( \frac{q}{2} \right) \times \cos \left( k - \frac{q}{2} \right) \cos \left( k' - \frac{q}{2} \right). \]  (4)

Three key new features appear in (4):

(a) it can be of either sign when \(k \neq k'\). This leads to a 'sign problem' in any Monte Carlo calculation (indeed, for any interaction \(M(k, q)\) with non-definite sign); we discuss this in the supporting material. The SSH model is thus representative of a large class of models in which non-diagonal couplings give a sign problem.

(b) multi-site hopping terms involving phonons generate terms in the polaron dispersion of form \(E(k) = E_0 - 2t_i^* \cos k - 2t_2^* \cos(2k) - ...\). Now for \(q\)-only dependent couplings, the nearest-neighbor hopping \(t_i^* \ll t_o\) is exponentially suppressed, and \(t_2^* \sim \frac{\omega_{ph}}{\omega_{ph}} e^{-\frac{Dq}{\omega_{ph}}} \) [18] is doubly suppressed, because each requires an intersite polaron cloud overlap. Here, however, an electron can hop from \(i-1 \rightarrow i \rightarrow i+1\), using only \(V\), to first create and then remove a phonon at site \(i\). The associated energy is \(t_2^* \propto \Delta t_{i,i+1} \Delta t_{i,i-1}/\omega_{ph}\) where \(\Delta t_{i,i-1} \sim \alpha\) is the phonon-induced change in the hopping. Since the phonon-induced displacement \(X_i\) increases one bond length while decreasing the other, \(\Delta t_{i,i+1} \Delta t_{i,i-1} < 0\), i.e., \(t_2^* \propto -\alpha^2/\omega_{ph}\) is negative, favoring a minimum in \(E(k) \sim -2t_2^* \cos(2k)\) at \(k = \pi/2\), consistent with our results for large \(\lambda\). This simple analysis indicates how the transition can occur. Of course, higher order terms must also be considered; and a transition like this, signalled by the change in the \(k_{\text{GS}}\), is certainly not guaranteed for all \(k-q\)-dependent couplings (thus the Edwards model in the large \(\lambda\) limit also has a dominant \(t_2^*\) term of similar origin; but \(t_2^* > 0\), and \(k_{\text{GS}} = 0\) for all \(\lambda\) [4]).

(c) Finally, consider the limit \(\omega_{ph} \rightarrow \infty\) for a fixed \(\lambda, t_o\). The phonon propagator tends to its static limit: \(D(q, \omega) = -2\omega_{ph}/(\omega_{ph}^2 - \omega^2) \rightarrow \tilde{D} = -2/\omega_{ph}\). The polaron propagator is then dominated by the 2nd-order correction in \(V\), scaling like \(\alpha^2/\omega_{ph} \sim \lambda t_o\) (higher order corrections \(\sim \alpha^{2n}/\omega_{ph}^{n-1} \sim t_o \lambda^{n-1} (\frac{\omega_{ph}}{\omega_{ph}})^{n-1} \rightarrow 0\). Thus, to lowest order in \(\omega_{ph}\) we get from Eq. 4 that

\[
E(k) = -2t_o \cos k + \frac{1}{2N^{1/2}} \tilde{D} \sum_q |M(k, q)|^2
= -2t_o \cos k - \frac{2\lambda t_o}{\sqrt{N}} \sum_q [\sin(k + q) - \sin k]^2 \]  (5)

We see that the dispersion curvature \([d^2 E(k)/dk^2]_{k=0} = 4t_o(1/2 - \lambda)\) at \(k = 0\). Thus, in the large \(\omega_{ph}\) limit, the effective mass diverges for \(\lambda_c(\omega_{ph} \rightarrow \infty) = 1/2\). Fig. 4 shows it converges very slowly to this limit.

This discussion shows, at least for sufficiently large \(\omega_{ph}\), that there must be a critical coupling strength \(\lambda_c\) at which \(k_{\text{GS}}\) leaves zero. For small \(\omega_{ph}\) the existence of a critical point is less clear, because the higher order diagrams can have arbitrary sign; but it is what we find here for all \(\omega_{ph}\) studied [19]. Note, however, that for \(\omega_{ph} < 0.3\), the average number of phonons \(N_{ph}\) increases significantly, making numerical simulations very difficult. The MA method is also questionable in this limit.

One is tempted to call this \(T = 0\) transition a 'quantum phase transition'. However this is not correct, because any phase transition must involve the cooperative behaviour of an infinite set of degrees of freedom; but here the number of phonons \(N_{ph}\) in the polaronic cloud always remains small. Of course with a macroscopic number of polarons in the system, we would see non-analyticity in bulk properties like \(dE_{\text{GS}}(\lambda)/d\lambda\); but a small number of polarons will be invisible in any thermodynamic property. Thus we simply assert the existence of a non-analyticity, as a function of \(\lambda\), in the polaronic properties.
We see that polarons having a coupling to a bosonic field depending on both $k$ and $q$ behave in a fundamentally different way from the standard case with only $q$-dependent coupling. This suggests a large zoology of so far unexplored behavior in many physically relevant systems. Note how surprisingly different the polaronic properties are here. For example, for large $\lambda$, $m^*(\lambda)$ decreases, and $Z(k_{GS})$ remains quite large. We see that "standard polaronic behavior" is really just a feature of models like the Holstein and Frohlich model.

Experimental signatures of the new behavior - notably, the critical point - will clearly be invisible in any thermodynamic measurements. However the divergence of the effective mass should be easily detectable in transport measurements; the polaron mobility $\mu \sim 1/m^*$ goes to zero at the critical point. Thus in any system where the charge mobility is carried by the polarons, this critical point should be very obvious. It would also be interesting to do ARPES experiments [20], where polarons can be ejected directly from the insulating state, allowing direct measurement of $E(k)$ and $Z(k)$. Apart from polyaniline, various organic semiconductors are known to have important non-diagonal coupling to phonons [21], as do several dimerized Mott semiconducting oxides [22]; in some of these, the coupling can be varied somewhat by pressure. However, any quantitative theory for such experiments must also include the coupling to longitudinal phonons, electron-electron interactions, and inter-chain coupling.

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4. Other examples where polaronic physics is important include magnetism, where one studies spin polarons (see, e.g., P. Fazekas, "Electron Correlation and Magnetism". World Scientific, 1999), or in biophysics (see, e.g., A. Scott, Phys. Rep. 217, 1 (1992).

SUPPORTING MATERIAL

(i) Quantum Monte Carlo Methods: As discussed in the main text, a key feature of the problem addressed here is that any Quantum Monte Carlo (QMC) method is faced by a sign problem (whereas in the conventional polaron problem, with $q$-dependent coupling only, the imaginary time polaron Green’s function and self-energy are sign-definite). Since the sign change can only happen when the internal particle line changes momentum, we see that non-crossing diagrams are positive-definite and only a fraction of the crossing diagrams will have an overall negative sign. This is of course not as severe as the sign-alternating series found in the many-body fermionic case.

The Bold Diagrammatic Monte Carlo (BDMC) technique [1] is a sign-problem tolerant method for many-body problems. It is thus a natural fit to the SSH polaron, and is one of the four techniques used in this letter. It improves on the simpler Green’s Function Diagrammatic Monte Carlo (G-DMC) and Self-Energy Monte Carlo ($\Sigma$-DMC) methods. The former is a Monte Carlo sampling of the conventional Feynman diagram expansion for the Green’s function $G(k, \tau)$ where $k$ and $\tau$ are the momentum and imaginary time respectively. The $\Sigma$-DMC method also expands the self-energy $\Sigma(k, \tau)$ diagrammatically, but contains fewer terms, thus provid-
The guidance on how to approximate the equations of motions (which also determines what parts of the diagrams are discarded) comes from the variational meaning of MA. As a result, MA satisfies multiple spectral weight sum rules exactly. The Momentum Average Method: The Momentum Average (MA) approximation solves the equation of motion for the Green’s function \( G(k, \omega) = \langle 0 \mid c_k(\omega + i\eta - \mathcal{H})^{-1} c_k^\dagger \mid 0 \rangle \), where \( |0\rangle \) is the vacuum. As usual, this solution appears in the form of an infinite hierarchy of coupled equations of motion. The main idea behind MA is to simplify the coefficients multiplying the generalized Green’s function in these equations so that this infinite hierarchy can be solved (quasi)analytically. Because it is formulated in terms of equations of motion and because there is no truncation at any order, MA is a non-perturbative approximation. In diagrammatic terms, MA is equivalent to a sum of all diagrams for the self-energy expansion, after discarding exponentially small contributions (compared to what is kept) from each diagram. As a result, MA satisfies multiple spectral weight sum rules exactly.

The guidance on how to approximate the equations of motions (which also determines what parts of the diagrams are discarded) comes from the variational meaning of MA. Essentially, one makes some assumption about the nature of the bosonic cloud, and keeps only terms consistent with it. For the Holstein model, it is already a very good approximation to assume that all the bosons in the cloud are at the same site – however, there can be arbitrarily many such bosons, and the particle can be arbitrarily far from the cloud. Further systematic increases in the variational space lead to systematic improvements of the MA approximation. For more complicated models, one needs to allow the cloud to be more extended. In this work, we use the straightforward generalization to our ”SSH model” of the 3-site version of MA detailed for the Edwards model in Ref. [6].

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the phonon cloud has to be more extended to describe in an appropriate way the relevant physical processes (the extension increases as $\omega_{ph}$ decreases). In this work we use a cluster of 5 neighbouring sites (the cluster can be arbitrarily far from the electron). Moreover we include an additional pair of phonons located on any pair of lattice sites. In this way the scattering processes between the charge carrier and up to two phonons in $q$-space are exactly treated, so that the weak coupling limit is optimally described. The main advantage of the LPBED method is that it is possible to calculate not only the self energy of the quasiparticle but any correlation function.
