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 λ, 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 λ_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 (e.g., in angle-resolved photoemission spectroscopy 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 [2–4], they exemplify in quantum field theory the passage from weak to strong coupling in a nontrivial model of a single particle coupled to a bosonic field [5]. The first serious nonperturbative studies by Feynman [6] of the Frohlich polaron are now a classic, but only recently were accurate results established across the whole range of coupling strengths [7]. Since then, exact numerical studies have been made of, e.g., D-dimensional Holstein polarons in various lattice geometries, with $D = 1, 2, 3$ [8], of 3D Rashba-Pekar polarons with short-range interactions [9], of pseudo Jahn-Teller polarons [10], 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 λ. 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 nonzero 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 Letter we study a specific example of this general case. The particle-boson coupling is taken from the well-known Su-Schrieffer-Heeger (SSH) model, introduced to describe electrons in 1D 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 $\mathcal{H} = H_0 + V + H_{\mathrm{ph}}$, where

$$H_0 = -t_0 \sum_i (c_i^\dagger c_{i+1} + \mathrm{H.c.}) \equiv \sum_k \epsilon_k c_k^\dagger c_k \tag{1}$$

describes the hopping of electrons between sites, with band dispersion $\epsilon_k = -2t_0 \cos(k)$ ($c_i^\dagger$ creates an electron on site i, $c_k^\dagger$ creates a momentum state k). The term $H_{\mathrm{ph}} = \omega_{\mathrm{ph}} \sum_i b_i^\dagger b_i$ describes dispersionless phonons ($b_i^\dagger$ creates a phonon on site i). The interaction is

$$\begin{aligned} V &= -\tilde{\alpha} t_0 \sum_i (\hat{X}_i - \hat{X}_{i+1})(c_i^\dagger c_{i+1} + \mathrm{H.c.}) \\ &= N^{-1/2} \sum_{k,q} M(k, q) c_{k+q}^\dagger c_k (b_{-q}^\dagger + b_q), \end{aligned} \tag{2}$$

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

$$\begin{aligned} M(k, q) &= 2i\alpha[\sin(k + q) - \sin(k)] \\ &= i(2\lambda\omega_{\mathrm{ph}} t_0)^{1/2}[\sin(k + q) - \sin(k)]. \end{aligned} \tag{3}$$

This interaction, with associated energy $\alpha = \tilde{\alpha} t_0 \times \sqrt{\hbar/2M\omega_{\mathrm{ph}}}$, describes the modulation of the hopping amplitude by phonons. We henceforth set $t_0 = 1$, and define two dimensionless parameters: the electron-phonon coupling parameter $\lambda = 2\alpha^2/(t_0\omega_{\mathrm{ph}}) = \langle |M(k, q)|^2 \rangle / (2t_0\omega_{\mathrm{ph}})$, where $\langle \cdot \rangle$ averages over the Brillouin zone, and the "adiabaticity" ratio ω_{ph}/t_0 ($\equiv \omega_{\mathrm{ph}}$ when $t_0 = 1$).

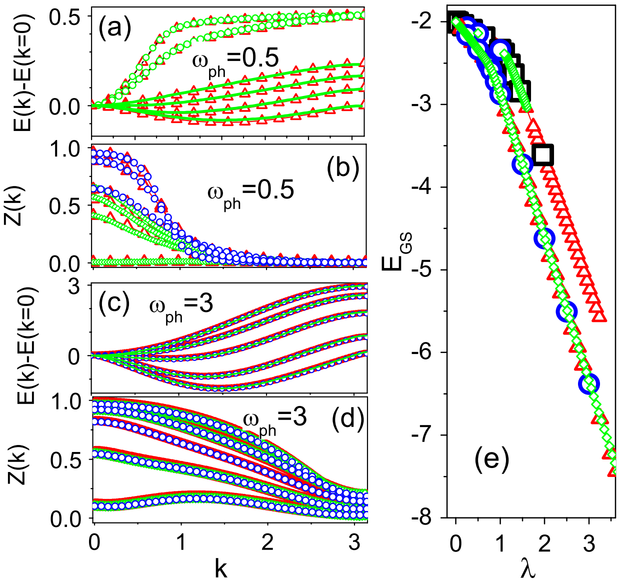

FIG. 1 (color online). The polaron dispersion relation $E(k) - E(k = 0)$ is shown in (a),(c), and the GS Z factors $Z(k)$ at momentum k are shown in (b),(d). Red triangles (blue circles) correspond to LPBED (BDMC) methods. In (a),(b), where $\omega_{\rm ph} = 0.5$, $\lambda = 0.25, 0.5, 1.0, 1.094, 1.21, 1.96$ (from top to bottom). In (c),(d), where $\omega_{\rm ph} = 3$, $\lambda = 0.25, 0.5, 1.0, 2.0, 4.0$ (from top to bottom). MA results are shown as green solid curves. In (e) the GS energy for $\omega_{\rm ph} = 0.5$ (upper line) and $\omega_{\rm ph} = 3$ (lower curve) is shown; triangles, rhombi, squares, and circles correspond to LPBED, MA, DMC, and BDMC methods, respectively.

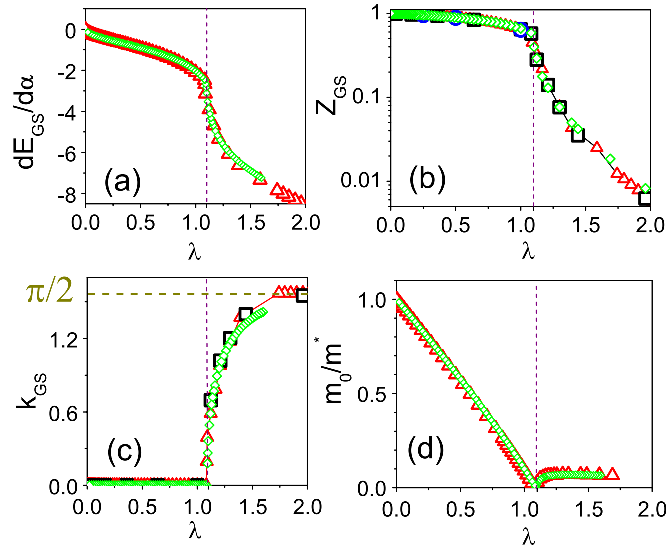

FIG. 2 (color online). (a) Derivative of the GS energy with respect to α, (b) Z factor of the GS, (c) wave vector of the GS, and (d) the ratio m_0/m^* of the bare and effective polaronic masses at $k_{\rm GS}$ for $\omega_{\rm ph} = 0.5$ (here $m_0 = 1/2t_0$). Red triangles, green rhombi, black squares, and blue circles correspond to LPBED, MA, DMC, and BDMC methods, respectively. The vertical dashed line indicates the critical coupling λ_c.

Results.—We treat this nonperturbative problem with the momentum average (MA) analytical approximation [13–15] and three different numerical techniques: the diagrammatic Monte Carlo (DMC) [7], the limited phonon basis exact diagonalization (LPBED) [16], and the bold diagrammatic Monte Carlo (BDMC) [17] methods. Applications of the first three methods to polaron problems are well documented. However, our implementation of the BDMC method for the SSH model contains several new elements, reviewed in the supplementary material [18].

In the following we display results as functions of λ in both the adiabatic regime (choosing $\omega_{\rm ph} = 0.5$) and the nonadiabatic regime (choosing $\omega_{\rm ph} = 3.0$). We begin with the quasiparticle dispersion $E(k)$ and renormalization factor $Z(k)$ [Figs. 1(a)–1(d)]. One sees immediately that whatever the adiabaticity, the minimum of $E(k)$ is at $k = 0$ for small λ but at finite k for large λ. At first glance, nevertheless, nothing unusual seems to happen to the ground state energy $E_{\rm GS}(\lambda)$ at the critical value λ_c, where $k_{\rm GS}$ first becomes nonzero [Fig. 1(e)]. In fact, the curves in Fig. 1(e) look quite similar to those for Holstein polarons.

However, there is actually a singularity at λ_c. Plots of the dimensionless derivative $dE_{\rm GS}(\lambda)/d\alpha$ [Fig. 2(a)], the overlap $Z_{\rm GS}(\lambda)$ between the ground state at finite λ and the uncoupled ground state [Fig. 2(b)], the momentum $k_{\rm GS}(\lambda)$ for which $E(k)$ is minimized [Fig. 2(c)], and the renormalized effective polaron mass $m^*(\lambda) = [\partial^2 E(\lambda)/\partial k^2]^{-1}|_{k=k_{\rm GS}}$ [Fig. 2(d)] all show a sharp transition at $\lambda = \lambda_c(\omega_{\rm ph})$ (see Fig. 2 for $\omega_{\rm ph} = 0.5$, Fig. 3 for $\omega_{\rm ph} = 3$). At this singularity, the polaronic mass $m^*(\lambda)$ diverges, with corresponding jumps in the first derivatives $dk_{\rm GS}(\lambda)/d\lambda$ and $dZ_{\rm GS}(\lambda)/d\lambda$, and in $d^2E_{\rm GS}(\lambda)/d\lambda^2$. However, the average number of phonons $N_{\rm ph}(\lambda)$ in the polaronic

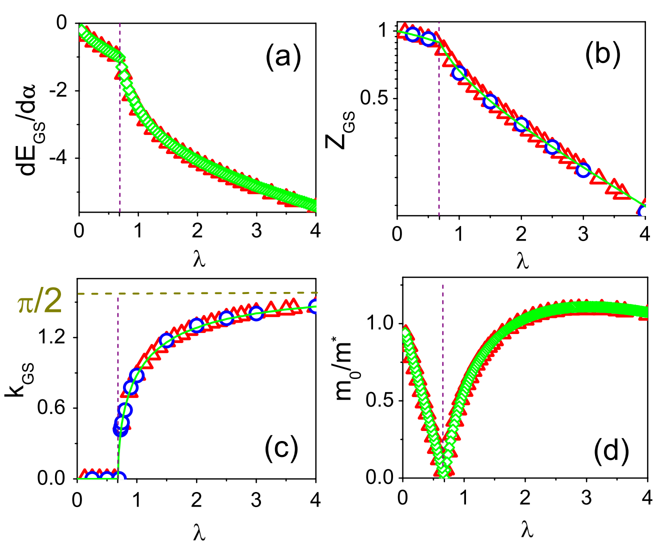

FIG. 3 (color online). The same as Fig. 2 but for $\omega_{\rm ph} = 3$. MA results in (b),(c) are shown as green solid lines.

polarization cloud does not diverge at λ_c (although it is presumably still singular); $N_{\rm ph}(\lambda_c) < 15$ for all values of the adiabaticity parameter $\omega_{\rm ph}$ checked so far. Note also how λ_c varies with $\omega_{\rm ph}$ (Fig. 4), initially increasing for small $\omega_{\rm ph}$, but then falling to the asymptotic value $\lambda_c \to 1/2$ in the instantaneous phonon limit $\omega_{\rm ph} \to \infty$. 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.

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 nondiagonal in site index. Thus phonons cause the bandwidth to fluctuate, and can by themselves generate hopping between sites. The lowest-order process contributing to $E_{\rm GS}$ 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 phonon of momentum q; we have

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

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 nondefinite sign); we discuss this in the supplementary material [18]. The SSH model is thus representative of a large class of models in which nondiagonal couplings give a sign problem.

(b) Multisite hopping terms involving phonons generate terms in the polaron dispersion of the form $E(k) = E_0 - 2t_1^* \cos k - 2t_2^* \cos(2k) - \cdots$. Now for q-only dependent couplings, the nearest-neighbor hopping $t_1^* \ll t_0$ is exponentially suppressed, and $t_2^* \sim \frac{t_0^2}{\omega_{\rm ph}} e^{-4\lambda t/\omega_{\rm ph}}$ [19] is doubly suppressed because each requires an intersite polaron cloud overlap. Here, however, an electron can hop from $i-1 \to i \to i+1$, using only V, to first create and then remove a phonon at site i. The associated energy is $t_2^* \propto \Delta t_{i,i+1}\Delta t_{i,i-1}/\omega_{\rm ph}$ where $\Delta t_{i,i-1} \sim \alpha$ is the phonon-induced change in the hopping. Since the phonon-induced displacement $\hat{X}_i$ increases one bond length while decreasing the other, $\Delta t_{i,i+1}\Delta t_{i,i-1} < 0$, i.e., $t_2^* \propto -\alpha^2/\omega_{\rm ph}$ is *negative*, favoring a minimum in $E(k) \sim -2t_2^* \cos(2k)$ at $k = \pi/2$, consistent with our results for large λ. This simple analysis indicates how the transition can occur. Of course, higher order terms must also be considered, and a transition like this, signaled by the change in $k_{\rm GS}$, is certainly not guaranteed for all k-q-dependent couplings (thus the Edwards model in the large λ limit also has a dominant t_2^* term of similar origin, but $t_2^* > 0$, and $k_{\rm GS} = 0$ for all λ [15]).

(c) Finally, consider the limit $\omega_{\rm ph} \to \infty$ for a fixed λ, t_0. The phonon propagator tends to its static limit: $D(q, \omega) = -2\omega_{\rm ph}/(\omega_{\rm ph}^2 - \omega^2) \to \tilde{D} = -2/\omega_{\rm ph}$. The polaron propagator is then dominated by the 2nd-order correction in V, scaling like $\alpha^2/\omega_{\rm ph} \sim \lambda t_0$ [higher order corrections $\sim \alpha^{2n}/\omega_{\rm ph}^{n-1} \sim t_0 \lambda^{n-1}(\frac{t_0}{\omega_{\rm ph}})^{n-1} \to 0$]. Thus, to lowest order in $\omega_{\rm ph}^{-1}$ we get from Eq. (4) that

$$\begin{aligned} E(k) &= -2t_0 \cos k + \frac{1}{2N^{1/2}}\tilde{D}\sum_q |M(k,q)|^2 \\ &= -2t_0 \cos k - \frac{2\lambda t_0}{\sqrt{N}}\sum_q [\sin(k+q) - \sin k]^2. \end{aligned} \quad (5)$$

We see that the dispersion curvature $[d^2E(k)/dk^2]_{k=0} = 4t_0(1/2 - \lambda)$ at $k = 0$. Thus, in the large $\omega_{\rm ph}$ limit, the effective mass diverges for $\lambda_c(\omega_{\rm ph} \to \infty) = 1/2$. Figure 4 shows it converges very slowly to this limit.

This discussion shows, at least for sufficiently large $\omega_{\rm ph}$, that there must be a critical coupling strength λ_c at which $k_{\rm GS}$ leaves zero. For small $\omega_{\rm ph}$ 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 $\omega_{\rm ph}$ studied [20]. Note, however, that for $\omega_{\rm ph} < 0.3$ the average number of phonons $N_{\rm ph}$ 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 behavior of an infinite set of degrees of freedom, but here the number of phonons $N_{\rm ph}$ in the polaronic cloud always remains small. Of course with a macroscopic number of polarons in the system, we would see nonanalyticity in bulk properties like $dE_{\rm GS}(\lambda)/d\lambda$, but a small number of polarons will be invisible in any thermodynamic property. Thus we simply assert the existence of a nonanalyticity, as a function of λ, in the polaronic properties.

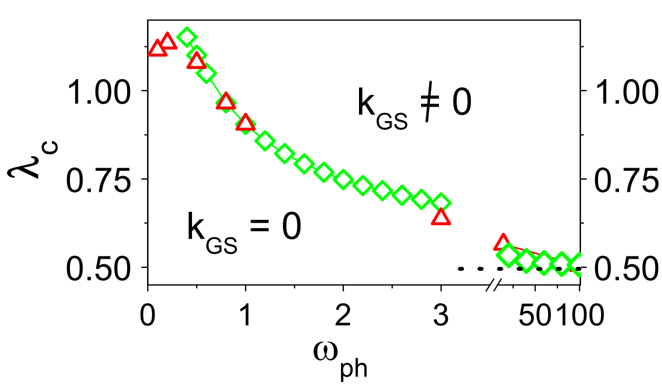

FIG. 4 (color online). Phase boundary dividing GSs with zero and nonzero momentum. Green squares and red triangles refer to MA and LPBED methods. The horizontal dotted line refers to the instantaneous limit $\lambda_c^{\omega_{\rm ph}\to\infty} = 1/2$.

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 λ, $m^*(\lambda)$ *decreases* and $Z(k_{\rm GS})$ 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 angle-resolved photoemission spectroscopy experiments [21], where polarons can be ejected directly from the insulating state, allowing direct measurement of $E(k)$ and $Z(k)$. Apart from polyacetylene, various organic semiconductors are known to have important nondiagonal coupling to phonons [22], as do several dimerized Mott magnetic semiconducting oxides [23]; 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 interchain coupling.

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