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Self-Trapping of Polarons in the Rashba-Pekar Model

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We performed quantum Monte Carlo study of the exciton-polaron model which features the self-trapping phenomenon when the coupling strength and/or particle momentum is varied. For the first time accurate data for energy, effective mass, the structure of the polaronic cloud, dispersion law, and spectral function are available throughout the crossover region. We observed that self-trapping can not be reduced to hybridization of two states with different lattice deformation, and that at least three states are involved in the crossover from light- to heavy-mass regimes.

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Properties of particles strongly coupled to their environment are of importance in many fields of physics and are attracting constant attention given extreme diversity of what may be called a "particle," an "environment," and how they interact with each other. In most general terms, self-trapping (ST) means a dramatic transformation of particle properties when system parameters are slightly changed. Landau[1] showed that the "trapped" (T) particle state with strong lattice deformation around it and the weakly perturbed "free" (F) particle state may have the same energy at some critical value of the coupling strength, α_c.

Of course, the resonance between F and T states is not infinitely sharp since the matrix element hybridizing them is non-zero, i.e. ST phenomenon is a crossover, rather than a transition, and all polaron properties are analytic in α—see Ref. 2 for an explicit proof. This theorem makes the notion of ST rather vague since there is always some admixture of one state in another. Moreover, it challenges the adopted opinion that only two, namely F and T, states are in competition. If there are more than two states within the energy scale of the hybridization matrix element then all of them are mixed and the F-T classification fails. In fact, the two-states assumption on which the current theory is based is not supported by experiments and rather complex spectra are usually observed instead[3,4].

According to the standard criterion[5,6], ST takes place if there is a barrier U_B in the adiabatic potential between the bare-particle and polaron states. It occurs, almost by definition, in the intermediate coupling regime where perturbation theory is not applicable. Hence, the existence of a barrier—if the very notion of the adiabatic potential is not ill defined—and the ST phenomenon can be addressed only by an exact method, because in the intermediate coupling regime an analytic solution is hardly available, and even sophisticated variational treatments often give misleading results[7].

In this Letter, we consider a typical model in which particle couples to the environment of gapped dispersionless optical phonons. For this model it is possible to define ST in a mathematically rigorous way and proceed with its quantitative study. We show how various particle properties (energy, effective mass, dispersion law, and the structure of the polaronic cloud) change between weak- and strong-coupling limits, and provide detailed information about ST of polarons, which is not based on any approximations. Besides, we show that there are at least three states involved in mixing in the critical region and, thus commonly accepted concept of only F and T states mixing at α_c appears to be oversimplified. In fact, we are not aware of any other numerical study testing how accurate are existing treatments of the ST problem.

The Hamiltonian of the system consists of the free-particle term (we consider continuum three-dimensional case with dispersion relation $\varepsilon(\mathbf{k}) = k^2/2m$)

$$H_{\rm e} \;=\; \sum_{\mathbf{k}} \varepsilon(\mathbf{k})\, a^\dagger_{\mathbf{k}} a_{\mathbf{k}} \;, \qquad (1)$$

the Hamiltonian of the phonon bath

$$H_{\rm ph} \;=\; \sum_{\mathbf{q}} \omega_q\, b^\dagger_{\mathbf{q}} b_{\mathbf{q}} = \omega_0 \sum_{\mathbf{q}} b^\dagger_{\mathbf{q}} b_{\mathbf{q}} \;, \qquad (2)$$

and the standard density-displacement interaction[8]

$$H_{\text{e-ph}} \;=\; \sum_{\mathbf{k},\mathbf{q}} V(\mathbf{q})\, \left(b^\dagger_{\mathbf{q}} - b_{-\mathbf{q}}\right)\, a^\dagger_{\mathbf{k}-\mathbf{q}} a_{\mathbf{k}} \;. \qquad (3)$$

In Eqs. (1-3), $a_{\mathbf{k}}$ and $b_{\mathbf{q}}$ are the particle and phonon annihilation operators in momentum space, correspondingly.

Our study is based on the quantum Monte Carlo simulation of the polaron Green function in imaginary time at $T = 0$ and subsequent analytic continuation to the real frequencies[9,10,11]. The method suggested in Refs.[9,10,11] is free from approximations and systematic errors. It is particularly suited for the study of ST problem where several δ-peaks are expected below the spectral continuum in the Lehman expansion

$$S^{(\mathbf{k})}(\omega) \;=\; \sum_{\nu} \delta(\omega - E_\nu(\mathbf{k}))\; |\langle \nu | a^\dagger_{\mathbf{k}} | \text{vac}\rangle|^2 \;. \qquad (4)$$

Here $\{|\nu\rangle\}$ is a complete set of eigenstates of H in the momentum sector $\mathbf{k}$, i.e. $H\,|\nu(\mathbf{k})\rangle = E_\nu(\mathbf{k})\,|\nu(\mathbf{k})\rangle$.

Separating stable quasiparticle states (labeled by index i) from continuum, we rewrite Eq. (4) as

$$S^{(\mathbf{k})}(\omega) \;=\; \sum_i Z_i^{\mathbf{k}}(0)\delta\left(\omega - E_i^{(\mathbf{k})}\right) + \int_{\omega_c} d\omega\, s^{(\mathbf{k})}(\omega)\;, \tag{5}$$

where $Z_i^{\mathbf{k}}(0)$ and $E_i^{(\mathbf{k})}$ are Z-factors and energies of stable states, and the continuum threshold is given by $\omega_c = E_0^{(\mathbf{k=0})} + \omega_0$. Any state with $E > \omega_c$ is unstable against single- ($n = 1$) or multi-phonon ($n > 1$) emission process $E \to E_{\rm i}^{(\mathbf{p})} + n\omega_0$, where momentum $\mathbf{p}$ is selected only by the energy conservation law since phonons are dispersionless.

Speaking rigorously, by self-trapping one understands the existence of such a region in the parameter space of H where more than one stable polaron states, differing by the degree of polarization of the lattice, coexist. This definition implies three critical points in the coupling constant (keeping other parameters fixed for simplicity), $\alpha_{c1}(\mathbf{k}) < \alpha_c(\mathbf{k}) < \alpha_{c2}(\mathbf{k})$. The ST "transition point" α_c is understood as the point of avoided crossing between the two lowest polaron states. At this point the groundstate of the polaron is a hybrid of states with substantially different degrees of lattice polarization. The critical points α_{c1} and α_{c2} correspond to the appearance and disappearance of the extra stable state(s), respectively. [By definition, the energy difference $\Delta E^{(\mathbf{k})}(\alpha)$ between the ground and first stable excited state, which has its minimum at $\alpha_c(\mathbf{k})$, ought to be less than $\omega_c - E_0^{(\mathbf{k})}$.] Critical couplings introduced above are consistent with previous considerations[1,5,6] and have an advantage of being unambiguous even when the minimal gap $\Delta E^{(\mathbf{k})}(\alpha_c(\mathbf{k}))$ is not small[12].

A typical system that is believed to feature ST is the so-called Rashba-Pekar model[13,14] which describes Wannier exciton in the $1s$ state interacting with optical vibrations via electrostatic potential[6]

$$V(\mathbf{q}) = \gamma(\mathbf{q})\left\{\frac{1}{[1+(\xi_e a_B q)^2]^2} - \frac{1}{[1+(\xi_h a_B q)^2]^2}\right\}, \tag{6}$$

$$\gamma(\mathbf{q}) \;=\; i\,\left(2\sqrt{2}\alpha\pi\right)^{1/2}\, q^{-1}\;. \tag{7}$$

Here α is the standard dimensionless coupling constant, a_B is the Bohr radius, and $\xi_{e,h} = m_{e,h}/[2(m_e + m_h)]$ is given in terms of electron (m_e) and hole (m_h) masses, respectively. In this Letter, we focus on the parameters corresponding to the curve number 2 of Ref.[13], which is believed to describe ST in the strong-coupling regime. Setting the total bare mass of the exciton $m = m_e + m_h$, phonon frequency ω_0, electric charge, and Plank constant to unity, one finds that $m_e = 0.065$. The Bohr radius can be used to change the degree of adiabaticity in the model. Below we shall thoroughly consider an "almost adiabatic" case (i) with $U_B/\omega_0 = 2$ (which is realised for $a_B = 0.467$) and outline some peculiar features of the "nonadiabatic" situation (ii) with $U_B/\omega_0 = 0.5$ ($a_B = 0.934$). The critical coupling constants, determined within the approach of Ref.[13], are then $\alpha_c^{\rm ad} \approx 14.3$ and $\alpha_c^{\rm ad} \approx 7.2$, respectively.

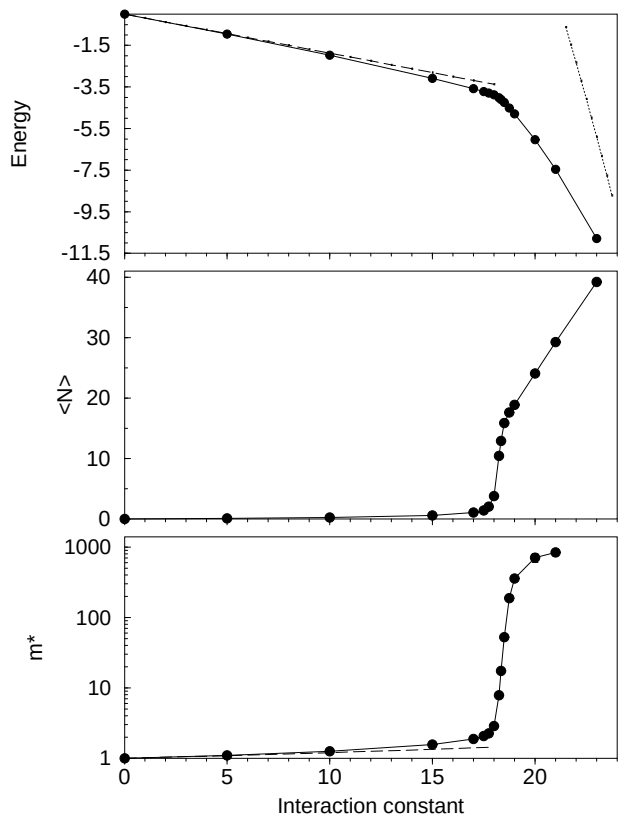

FIG. 1: The groundstate energy, average number of phonons, and effective mass as functions of α (points connected by solid lines). Relative statistic errors are less than 10^{-3} and 10^{-2} for the energy and $\langle N\rangle$, respectively. The relative statistic errors for the mass are of order 10^{-2} for $\alpha < 18.5$ and around 5×10^{-2} for larger coupling constants. Dashed lines show results of the perturbation theory while the dotted line corresponds to the strong-coupling limit.

In Fig. 1 we show how the groundstate properties ($\mathbf{k} = 0$) depend on the coupling strength. The groundstate energy, the effective mass m^*, and the average number of phonons in the polaronic cloud

$$\langle N\rangle = \langle \mathbf{k}=0|\sum_{\mathbf{q}} b_{\mathbf{q}}^\dagger b_{\mathbf{q}}\,|\mathbf{k}=0\rangle, \tag{8}$$

clearly indicate drastic changes around $\alpha_c \approx 18.35$. At this point the energy derivative changes very fast, and both $\langle N\rangle$ and m^* ungergo step-wise increase visible even on the logarithmic plot for m^*. In a narrow region between $\alpha = 17.5$ and $\alpha = 19$ the effective mass increases by two orders of magnitude. The remarkable fact is that at α_c the strong-coupling approach is still far from being accurate (dotted line[14] in the upper panel of Fig. 1). Besides, the adiabatic critical constant $\alpha_c^{\rm ad} \approx 21$[14] differs

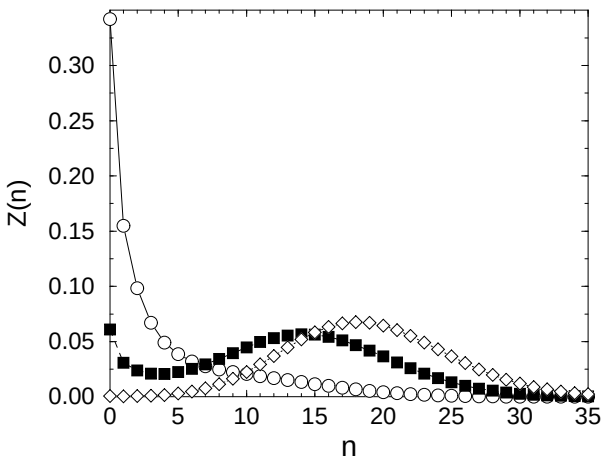

FIG. 2: Partial weights of n-phonon states in the polaron ground state ($\mathbf{k}=0$) at $\alpha = 18$ (circles), $\alpha = 18.35$ (squares), and $\alpha = 19$ (dimonds). Statistic errorbars of order 3×10^{-3} are less than the symbol size.

significantly from our value $\alpha_c \approx 18.35$. For the "nonadibatic" case (ii) the behavior of the groundstate properties is qualitatively the same, but quantitative deviations from the strong-coupling limit are larger.

Next, we study how the phonon cloud evolves throughout the ST critical region. Partial n-phonon contributions to the polaron ground state $Z_0^{\mathbf{k}}(n)$ are the probabilities of finding exactly n phonons in the cloud, and the average number of phonons introduced earlier, is just $\langle N \rangle = \sum_n nZ(n)$. Figure 2 shows $Z_0^{\mathbf{k}=0}(n)$ distributions at $\alpha = 18$ (below the crossover region), $\alpha = \alpha_c = 18.35$, and $\alpha = 19$ (trapped state). We see that the distribution at α_c has two peaks and is half-way between the two limiting cases. However, in the "nonadiabtic" case (ii) the structure with two maxima in $Z(n)$ is missing. Therefore, the peculiar behavior presented in Fig. 1 is a general feature of the ST phenomenon whereas the two-peak structure of the phonon distribution is specific for the adiabatic limit.

The spectral function around the critical point[15] reveals up to three stable excited states below the continuum threshold (see examples of the Lehman function in Fig. 3). We observe in Fig. 4 that three polaronic states (in the energy range comparable with the hybridization strength) participate in the ST crossover. We underline that all three states have large Z-factors (> 0.1). Therefore, for the given set of parameters more than two states are mixed at the crossover point and the standard picture of F-T hybridization at the tip of the ST crossover fails. One can speculate that extra stable states in the gap, which standard theory puts into the spectral continuum, are due to excited levels of highly nonlinear ST potential in the resonating region. However, this interpretation is essentially qualitative since the concept of adiabatic potential breaks down in the crossover region.

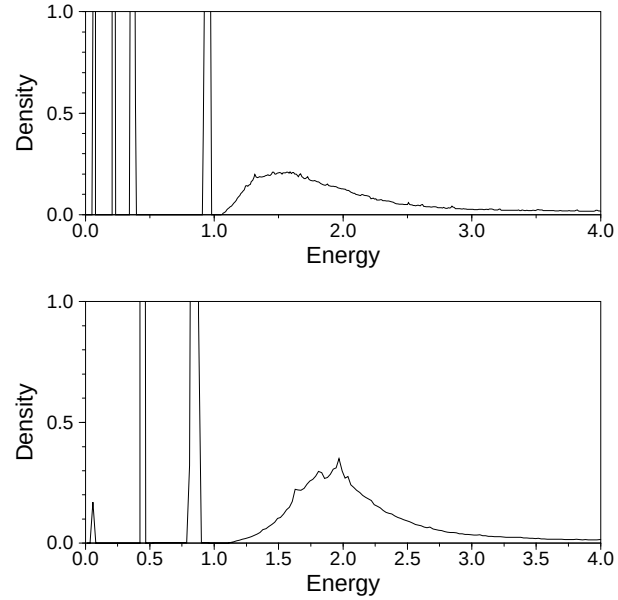

FIG. 3: The Lehman spectral function $S^{(\mathbf{k}=0)}$ at coupling constants $\alpha = 18.35$ (upper panel) and $\alpha = 18.75$ (lower panel).

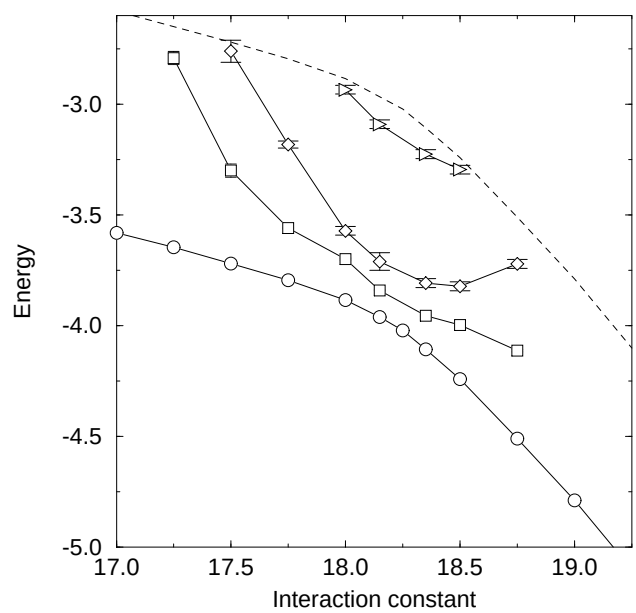

FIG. 4: Energies of the ground (circles) and excited stable states (squares, diamonds, and triangles) *vs* interaction constant. The dashed line is the threshold of incoherent continuum. Typical errorbars for the first, second, and third excited states are 10^{-2}, 3×10^{-2}, and 4×10^{-2}, respectively.

So far we have considered ST crossover at zero momentum, i.e., for the ground state. However, same considerations apply to finite momentum states, as long as $k < k_c$ where k_c is defined as the point where $E_0^{(\mathbf{k})} = \omega_c$ and the polaron spectrum has an end point[9,16]. For $\alpha < \alpha_c$ the

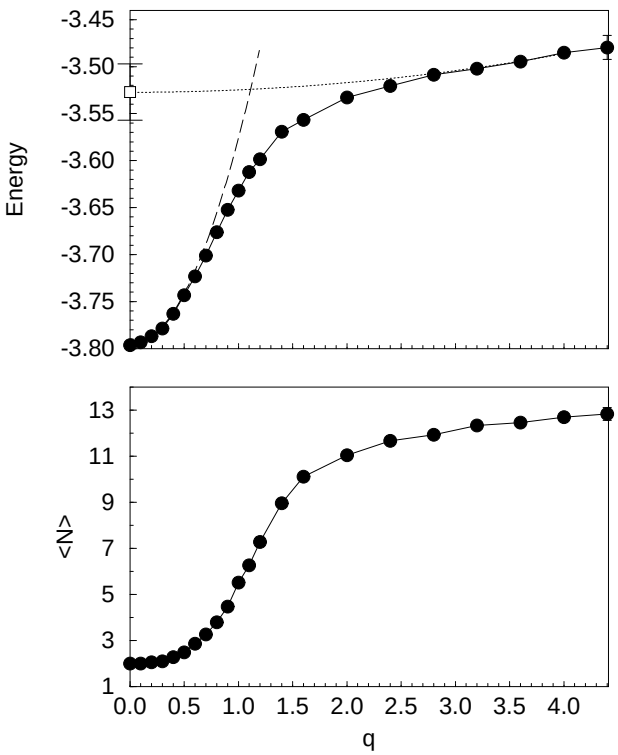

FIG. 5: The wave-vector dependence of energy and average number of phonons for $\alpha = 17.75$ (circles connected by solid lines). The errorbars are 3×10^{-3} and 10^{-2} for energy and $\langle N \rangle$, respectively. The dashed curve is the effective mass approximation $E^{(\mathbf{k})} = E_0 + \mathbf{k}^2/2m^*$ with $E_0(\alpha = 17.75) = -3.7946$ and $m^*(\alpha = 17.75) = 2.258$ obtained from direct Monte Carlo estimators. The dotted curve is the parabolic dispersion law fitted to the last four points in the energy plot with parameters $E_1(\alpha = 17.75) = -3.5273$ and $m_1^* = 195$. The open rectangle is the energy obtained from spectral analysis for the first excited state.

ground state is characterized by small lattice distortion and light effective mass and thus the dispersion law associated with this state rises steep with $\mathbf{k}$. On another hand, the dispersion curve of the excited heavy-mass state is nearly flat which means that light- and heavy-branches have to intersect at some wave vector. In Fig. 5 we plot the groundstate dispersion law and the average number of phonons in the ground state for $\alpha = 17.75$ which we interpret as the level crossing picture (see the figure caption). Notice the agreement between the spectral analysis at $\mathbf{k} = 0$ and the parabolic fit of the heavy mass branch.

By all accounts, the existence of more than one stable polaron state is a highly nontrivial qualitative property of the model whether the hybridization gap between these states is small, or not. Moreover, the ST crossover is not necessarily limited to hybridization of only two states. Recent studies of the Holstein polaron in 1D strongly support the universality of the latter statement since more than two stable states were found there as well[17].

We found that the dependence of E_0 and $\langle N \rangle$ (but not the structure of the cloud) on coupling can be used as an indirect indication of the self trapping phenomenon with a well-defined "transition point" (the point of minimal gap between the ground and first stable excited state). It was considered previously as a theorem, that ST may occur only in dimensions $d > 2$[5,6]; however, the definition of what has to be counted as a ST transition was not given in quantitative terms. We believe that it is more appropriate to use less "radical", but unambiguous, definition adopted in our paper. Recently, the second stable polaron state was found to exist for the Holstein polaron in a one-dimensional lattice[18] and in infinite dimension approximation[19].

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