Polaron Formation in One-Dimensional Quasiperiodic Systems

St. Pnevmatikos,[a] O. Yanovitskii,[b] Th. Fraggis, and E. N. Economou[c]
Research Center of Crete–Foundation for Research and Technology-Hellas, P.O. Box 1527, 711 10 Heraklion, Crete, Greece
(Received 12 August 1991)

We have studied through real-time numerical simulations the question of polaron formation by a single electron injected in a band consisting of fluctuating amplitude or mobility edge eigenstates. We found that polaron formation depends strongly on how close to a mobility edge the initial state is, on its average energy, and, to a much lesser degree, on its shape. In some instances novel highly excited long-lived localized polarons appear. In general, the time evolution exhibits an unexpected behavior; even for very long times, the electron does not seem to transfer much of its energy to the lattice.

PACS numbers: 71.38.+i

The question of polaron formation has been studied extensively as a ground-state problem in periodic (or uniform) systems [1]. However, there are two questions which have not yet received satisfactory answers: How polaron formation occurs near a mobility edge [2]? What is happening in the case where an electron is injected (or is photoexcited) well within an empty band in the presence of electron-phonon (el-ph) interaction? The usual answer to the second question is based on the assumption that the electron gradually transfers its energy to the lattice, reaches the bottom of the empty band, and then the question of polaron formation is again treated as a ground-state problem. In this Letter, we report results (some of them quite unexpected) based on a novel approach employing techniques borrowed from nonlinear physics, i.e., numerical simulations of the time evolution of an initial electronic state as it interacts with the lattice. Our approach can be easily generalized to treat the problem of two electrons interacting with each other and with the lattice. Thus, it offers itself for the study of bipolaron formation [3] especially in the difficult case where the electron and ion masses are of similar magnitude; this case may be relevant to narrow-band high-T_c superconductors [4] or the newly discovered fullerides [5].

The Hamiltonian describing our system is $H=H_e+H_l+H_{e\text{-}l}$. The electronic part, H_e, is given by

$$H_e=\sum_n \varepsilon_n|n\rangle\langle n| - V\sum_{nm}{}'|n\rangle\langle m|\,, \qquad (1)$$

$$\varepsilon_n=\varepsilon_0\cos(2\pi\sigma n),\quad V>0\,,$$

where the local orbital $|n\rangle$ is centered around site n ($\{n\}=1,\ldots,N$ form a 1D lattice with lattice constant a and periodic boundary conditions: $|N+1\rangle=|1\rangle$); σ is an irrational number taken as the "golden mean" $(\sqrt{5}+1)/2$; the prime in the summation denotes nearest-neighbor pairs only. The Hamiltonian (1) was chosen because it possesses Bloch eigenstates (for $\varepsilon_0=0$), mobility edge eigenstates (when $\varepsilon_0/V=2$), and all intermediate fluctuating extended eigenstates (when $0<\varepsilon_0/V<2$), giving thus the opportunity to study in a simple 1D model, the first of our questions. The disadvantage of Hamiltonian (1) is its 1D character [for $D\le 2$, as opposed to $D=3$, a localized (in the framework of the static Emin-Holstein [6] approximation) polaron is always the ground state no matter how weak the el-ph coupling is] and the fact that its spectrum is very anomalous [7].

The lattice part, H_l, is given by

$$H_l=\tfrac{1}{2}m\sum_n \dot{u}_n^2+\tfrac{1}{2}\kappa\sum_n (u_{n+1}-u_n)^2\,, \qquad (2)$$

where m is the ionic mass, u_n is the displacement, $\dot{u}_n$ is the velocity of ion n, and κ is the "spring constant." The lattice vibration part is treated classically [8]. The maximum eigenfrequency is $2\omega_0$, and the sound velocity is $c=\omega_0 a$, where $\omega_0=\sqrt{\kappa/m}\equiv t_l^{-1}$. The electron-lattice interaction is taken as a symmetrized deformation potential, i.e.,

$$H_{e\text{-}l}=\chi\sum_n |n\rangle\langle n|(u_{n+1}-u_{n-1})\,, \qquad (3)$$

where χ is the strength of the coupling ($2a\chi$ is the deformation potential). The strength of the coupling can also be characterized by the dimensionless quantity

$$\lambda=\chi^2/\kappa V \qquad (4)$$

which is similar to the λ which appears in the theory of superconductivity. Typical values for λ in metals are in the range 0.1 to 1.5.

Initially, the lattice is at rest and undeformed and the electron is placed in one of the following states: (a) in an eigenstate (corresponding to an eigenenergy near the bottom of the band or near its center or in between); (b) in a uniform state $[|\psi(0)\rangle=(1/\sqrt{N})\sum_n|n\rangle]$; (c) in a Gaussian wave packet with a standard deviation $\bar{\sigma}=3$ and initial energy $-1.949V$; (d) in the state $|\psi(0)\rangle=|n\rangle$ with n close to $[N/2]$ and energy close to zero; and (e) in the state $|\psi(0)\rangle=\sum c_n|n\rangle$ with $c_n=1/\sqrt{5}$, $-1/\sqrt{5}$, $1/\sqrt{5}$, $-1/\sqrt{5}$, $1/\sqrt{5}$, for five consecutive sites around the middle of the specimen, and energy close to zero. In our numerical work, we have taken V as our unit of energy and $t_l/t_e=81.8$, where $t_e=\hbar/V$ is the characteristic electronic time. We have chosen $t_0\equiv t_l(t_l/t_e)$ as our unit of time. The time integration is based on the fourth-order Runge-Kutta method with a step equal to $2\times10^{-6}t_0$. The size of the step was such that, during the numerical simulation, energy is conserved to an accuracy of 10^{-5} or better.

We classify our findings for the behavior of the polarons in three categories: (a) localized polarons, (b) large polarons, and (c) intermediate polarons. The third

category, which spans a rather wide range of parameter space, may or may not disappear for an infinite system and very long time lapse, $t \gg \tau$, where τ is the relaxation time. An approximate estimation for τ can be obtained using perturbation theory from the formula $\tau^{-1} = (2\pi/\hbar)\lambda h_l$, where h_l is the time average lattice energy per site, which is easily obtained from our calculations. If the electronic and lattice degrees of freedom were quasi-independent, one expects that equilibration of their respective temperatures would be established for $t \approx (10\text{–}100)\tau$. In all cases of highly excited electrons, we found that such equilibration was not even approached; instead, the lattice temperature either reached or approached a steady value orders of magnitude lower than the electronic temperature during a time lapse which in some instances exceeded 5000τ. This behavior strongly suggests that even in cases of weak electron-lattice interaction the electronic and lattice degrees of freedom cannot be considered as quasi-independent. Rather, as the polaron (even the large polaron) is formed (usually in steps), the polaron-lattice coupling is renormalized to smaller and smaller values, until, eventually, at least in some cases, it reaches a vanishingly small value.

To study the question of polaron behavior, we have examined the time development of (a) the electronic wave function, (b) the participation number $P = \sum |c_n|^4)^{-1}$, where $c_n = \langle n|\psi(t)\rangle$, (c) the electronic energy E_e, (d) the lattice energy $E_l \equiv H$ (both the kinetic and the potential part), (e) the interaction energy $E_{e\text{-}l} = \langle H_{e\text{-}l}\rangle$, and (f) the mean square displacement of the wave function $x^2(t) = \sum |c_n(t)|^2 (n - n_0)^2$, where $n_0 = \sum |c_n(0)|^2 n$.

In Fig. 1, we show results for $\varepsilon_0 = 0.5$ (weak departure from periodicity), $\lambda = 0.095$ (weak coupling), and a uniform initial state. The number of sites was chosen as $N = 377$ [which, being a Fibonacci number, makes the difference $\varepsilon_{N+1} - \varepsilon_1$ small ($=0.0035$)]. The average energy of the initial state is -2.001, which almost coincides with the bottom of the unperturbed band. Figure 1 shows clearly that a localized polaron is formed after about two units of time. [To exclude the possibility that this localization may be a finite-size effect, we have repeated the calculation for $N = 611$ (the next Fibonacci number).

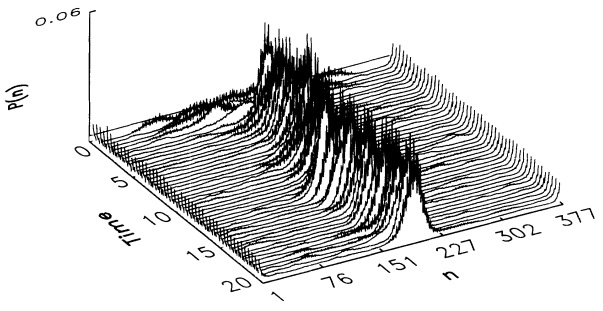

FIG. 1. Localized polaron formation from a uniform initial state for $N = 377$, $\varepsilon_0 = 0.5V$, and $\lambda = 0.095$. $P(n) \equiv |c_n(t)|^2$. The unit of time is $t_0 = 81.8 t_l$.

We found essentially the same results as in the $N = 377$ case.] Even after a polaron is formed, there are variations in its size with time, the most characteristic of which is the almost periodic pronounced contractions at $t = 2.5$, 7.5, 12, 16.5, etc., appearing as sharp peaks in the lattice energy and the electronic energy and sharp valleys in the interaction energy and the participation number. This breathing character of the localized polaron seems to be due to a coherent motion of the lattice where the peaks of the lattice potential energy coincide with the valleys in the lattice kinetic energy (the latter is on the average about 20% of the former). Polaron formation is associated with a decrease in P from the initial value $P = 377$ (or $P = 611$) to a quasiequilibrium value of $P \approx 50$ with values as low as 30 during the contraction periods. In the initial phase ($t < 2$) the localized polaron starts forming in a nonmonotonic way through successive contractions, each one followed by a short expansion apparently because of local overshooting. Note also that the electronic energy increases on the average, while the interaction energy, being negative, compensates for the variation of both E_e and E_l. Similar results were obtained for the periodic case ($\varepsilon_0 = 0$) with the same λ ($=0.095$) and a uniform initial state (which in this case is the ground state of the unperturbed electronic system). The main difference is that in the $\varepsilon_0 = 0$ case, all quantities have a much smoother time evolution.

Increasing the initial value of the energy of the electron requires larger values of either λ or ε_0 to obtain a localized polaron. In the case where the initial electronic energy is near the center of the band, ε_0 must exceed unity in order to obtain a localized polaron for any reasonable value of λ. As the mobility edge is approached, localized polarons appear for quite small values of λ. The center-of-the-band localized polarons consist usually of more than one rather sharp peak, each peak sometimes being independently mobile, thus giving rise to local excitations of fractional charge. These novel, highly excited, apparently stable (no sign of disintegration was observed even for very long time lapses of the order of $3 \times 10^4 t_l$) localized polarons are reminiscent of solitons in polyacetylene, only richer in behavior.

It is worthwhile to point out that, in spite of the apparently chaotic noisy behavior of the various quantities, long time scales appear either dependent or independent of N. Furthermore, if $\psi(0)$ is an eigenstate the transfer of energy to the lattice is much larger than in the other cases in spite of the fact that for $t \simeq 3t_l$, $\psi(t)$ looks equally extended for all initial states of the same energy. This suggests that the system retains some memory of the initial state for a very long time ($t \gtrsim 3 \times 10^4 t_l$).

A case of large polaron was obtained starting from a single-site, type-(d), initial state with energy near the center of the band ($E_e = 0.016$), $\varepsilon_0 = 1$, and $\lambda = 0.86$ (rather strong electron-lattice coupling). The initial wave packet expands at the beginning ($t \lesssim 3t_l$) ballistically with a mean square displacement $x^2 \sim t^a$, where $a \approx 2$.

The maximum value of $(x^2)^{1/2}$ is about 120; the value of P remains almost constant at 185 ± 15 (for $0.05 < t < 380$) with no tendency for reduction in spite of the extremely long running time (about $3 \times 10^4 t_l$). The lattice energy has not saturated, although dE_l/dt is decreasing on the average in a steplike way. The lattice kinetic and potential energies are equal to each other indicating an internal lattice thermalization. The electron-lattice interaction energy is very small (about -0.001). In spite of the very long time run, only a very small fraction of the electron energy has been transferred to the lattice (0.095). The "final" electronic energy of -0.08 corresponds to an electronic temperature of the order of 30000 K, while the final lattice energy of 0.095 corresponds to a lattice temperature of 2.9 K. Extrapolating linearly the H_l vs t curve, we conclude that thermal equilibrium between lattice and electronic degrees of freedom will require times of the order of $10^6 \tau$, if it is going to be achieved at all. A quite similar behavior was observed even for a much smaller value of λ ($\lambda \approx 0.024$) and larger values of ε_0 ($\varepsilon_0 = 1.5$) and for the Gaussian wave packet. As in the previous case, there is no sign of formation of a localized polaron. In this case, in spite of the fact that $\tau = 63$, it seems that a steady state has been reached for $t \approx 140$ with $P \approx 125$, $E_e \approx -1.961$, $H_l \approx 0.012$, and $H_{e\text{-}l} \approx -0.0003$. Again the effective electronic temperature is of the order of 5000 K while the effective lattice temperature is 0.37 K. Repeating the calculations for $N = 611$, we found that the steady state is approached at a slower rate and that the final P increases proportionally to N. This shows that our large polarons are either extended or have a localization length much larger than 611. The above results support the idea that even large polarons are formed before much of the electronic energy is transferred to the lattice and that polarons have a very (or vanishingly) small coupling to the lattice. It is not clear whether or not this important result (according to which only a small fraction of the energy even of a highly excited electron is transferred to the lattice) is valid for 2D or 3D systems. In any case, it can possibly be checked experimentally by photoexciting electrons to the middle of the conduction band in quantum wires at very low temperatures.

We observed many cases of intermediate-nature polarons in which a rather mobile and deformable peak in the wave function coexists with what seems to be a more or less uniform background. We have termed these cases intermediate polarons. In most of these cases, although we followed the temporal evolution for a long time, it was not possible to find out whether eventually the polaron would become localized or large because periods of clear localized polaron formation are followed by periods of rather large polarons or vice versa. Increasing the length from $N = 377$ to $N = 611$ did not change this intermediate behavior. One such case is shown in Fig. 2, where a temporary localized polaron, associated with a strong valley in the participation ratio, is dissolved to a resonancelike state. Details of these very interesting intermediate cases will be presented elsewhere.

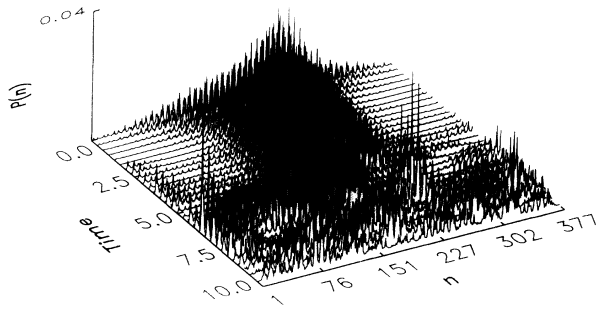

FIG. 2. Time evolution of an intermediate-type polaron from an initial eigenstate with $E_e = -1.942$, for $N = 377$, $\varepsilon_0 = 1$, and $\lambda \approx 0.38$. A temporary localized polaron is formed and then it dissolves to a resonancelike state.

At the critical point $\varepsilon_0 = 2$ the behavior of an initial single-site, type-(d), state with average energy equal to 0.034 seems to be the following: For no coupling ($\lambda = 0$), the motion appears to be diffusive (we find that $x^2 \sim t^\alpha$ with $\alpha \approx 0.9 \pm 0.1$). For a very weak coupling ($\lambda = 0.024$), the motion at the beginning $t \lesssim 4 t_l$ is diffusive with the same $\alpha \approx 1$ and then becomes subdiffusive ($\alpha \approx 0.6$) with strong fluctuations in x^2. A steady state appears to set in for $t > 5$ with $P \approx 110 \pm 10$, $H_l \approx 0.006$, $E_e \approx 0.028$, and $H_{e\text{-}l} \approx 0.0003$. Note that P in the absence of coupling, saturates at about 80. The observed increase of P for $\lambda \approx 0.024$ may be attributed to a "phonon-assisted" smoothing of the very fragmented mobility edge states. In the present case of very weak λ (≈ 0.024) our data do not allow any conclusion regarding the ultimate localization of the polaron, although it is conceivable that for long enough specimen and long times the x^2 vs t will saturate indicating thus that localization is setting in. As we increase the coupling λ to 0.095 (keeping all other relevant quantities the same), again x^2 vs t starts diffusively and then the motion becomes subdiffusive with $\alpha \approx 0.6$; finally x^2 vs t saturates. Further increase of λ to $\lambda = 0.38$ confirms the picture that already emerged. The transition from diffusive ($\alpha \approx 1$) to subdiffusive ($\alpha \approx 0.6$) motion occurs at $t \approx 3 t_l$ and the saturation value of both x^2 (57) and the participation number ($P \simeq 25 \pm 5$) become even smaller, demonstrating a lattice-induced localization. In conclusion, at the mobility edge an extremely weak coupling is enough to create a localized polaron even for the most difficult to localize, type-(d), state.

In Fig. 3, we summarize our findings by indicating the regions in the $\sqrt{\lambda}$ vs ε_0 plane where the polarons are localized, or intermediate, or large. In Fig. 3(b), we show results for initial electronic energy near the bottom of the band (Gaussian wave packet; similar results were obtained for the uniform initial state and for eigenstates of similar energy). In Fig. 3(a), we show results for initial energy near the center of the band [single-site, type-(d),

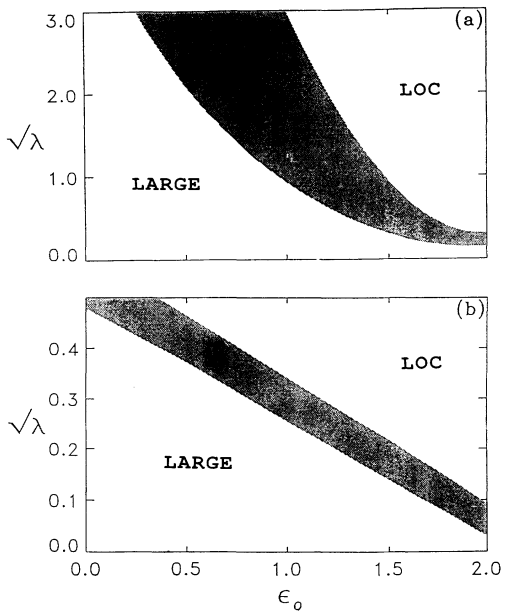

FIG. 3. Separation of ε_0-$\sqrt{\lambda}$ plane into regions of large, intermediate (I), and localized (LOC) polarons for initial energy (a) at the center of the band, $E_e=0$, and (b) near the bottom of the band, $E_e=-1.949$.

states; similar results were obtained for the type-(e) state and for eigenstates of similar energy]. It must be stressed that there is an ambiguity in finding the boundaries between the localized and the intermediate states and between the latter and the large states. Furthermore, for an infinite system and infinite time lapse, it is probable that part of the intermediate (or even the large) region may become localized. It must be pointed out that besides the obvious criteria for deciding whether or not a polaron is localized (i.e., shape of the eigenfunction, participation number, independence of N, time evolution of x^2), a clear sign of localization is obtained if $E_{e,f}-E_{e,i}>0$ and $|E_{e\text{-}l,f}|>E_{l,f}$, where the subscript i denotes initial and the subscript f denotes final state. Similarly, if $|E_{e\text{-}l}| \ll E_{l,f}$, this constitutes a strong indication for extended polarons.

From Fig. 3, we see that near or at the mobility edge a localized polaron is formed even for extremely weak electron-lattice interaction for all types of initial electronic states (even for those with average energy near the center of the band). This feature must be probably attributed to the very fragmented nature of the eigenstate near a mobility edge rather than to the pointlike nature of the unperturbed spectrum, since the latter will be "smoothed out" in the presence of the interaction. Thus, the present work supports the results of the approximate approach followed in Ref. [2] and restores, through the polaron formation, the possibility of a minimum metallic conductivity [2] advocated by Mott [9]. Another important result of our study is also shown in Fig. 3, where a much stronger electron-lattice interaction is needed in order to form a localized polaron as we move towards the center of the band and away from the mobility edge.

In conclusion, we point out that the important findings of our novel approach are the following: (i) As the mobility edge is approached an extremely weak coupling suffices to create localized (although not necessarily atomic size) polarons. This result is likely to be valid in 3D systems as well, in view of the results of Ref. [2]. (ii) Close to the mobility edge or for very large coupling, localized, high energy, stable polarons of fractional charge appear. (iii) As polarons are gradually created in steps, the effective polaron-lattice coupling is reduced possibly to vanishingly small value even for large polarons. (iv) Interesting and unexpected time evolution appears involving short-scale noisy behavior, long-time scales, and long-term memory. It is not clear whether the result (iii) above is a peculiar feature of the present model or is of more general validity.

We acknowledge financial support by CEC through the ESPIRIT basic research Grant No. 3041 and NATO Grant No. RG 769/87.

---

[a] Deceased.

[b] Also at Institute of Theoretical Physics, Academy of Sciences of the Ukrainian SSR, Kiev, Ukraine.

[c] Also at the Department of Physics, University of Crete, Crete, Greece.

[1] For a recent review, see, e.g., B Gerlach and H. Löwen, Rev. Mod. Phys. **63**, 63 (1991).

[2] Morrel H. Cohen, E. N. Economou, and C. M. Soukoulis, Phys. Rev. Lett. **51**, 1202 (1983).

[3] Morrel H. Cohen, E. N. Economou, and C. M. Soukoulis, Phys. Rev. B **29**, 4496 (1984); **29**, 4500 (1984).

[4] See, e.g., R. Micnas, J. Ranninger, and S. Robaszkiewicz, Rev. Mod. Phys. **62**, 43 (1990).

[5] A. F. Hebard *et al.*, Nature (London) **350**, 500 (1991); M. J. Rosseinsky *et al.*, Phys. Rev. Lett. **66**, 2830 (1991); K. Tanigaki *et al.*, Nature (London) **352**, 222 (1991); S. P. Kelty *et al.*, Nature (London) **352**, 223 (1991).

[6] D. Emin and T. Holstein, Phys. Rev. Lett. **36**, 323 (1976).

[7] See, e.g., B. Simon, Adv. Appl. Math. **3**, 463 (1982); J. B. Sokoloff, Phys. Rep. **126**, 189 (1985).

[8] This classical approximation may contribute to the immobilization of the polaron since, in contrast to the quantum lattice vibrations, in the present scheme there is the possibility of vanishing lattice kinetic energy.

[9] See, e.g., N. F. Mott and E. A. Davis, *Electronic Processes in Non-Crystalline Materials* (Clarendon, Oxford, 1979); N. F. Mott, in *Localisation 1991*, edited by K. A. Benedict and J. T. Chalker (Institute of Physics Publishing Ltd., Bristol, 1991).

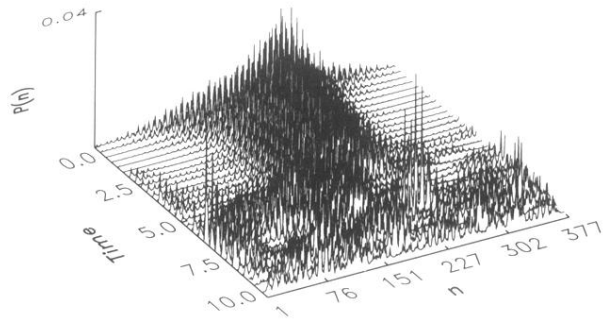

FIG. 2. Time evolution of an intermediate-type polaron from an initial eigenstate with $E_c = -1.942$, for $N = 377$, $\varepsilon_0 = 1$, and $\lambda \approx 0.38$. A temporary localized polaron is formed and then it dissolves to a resonancelike state.

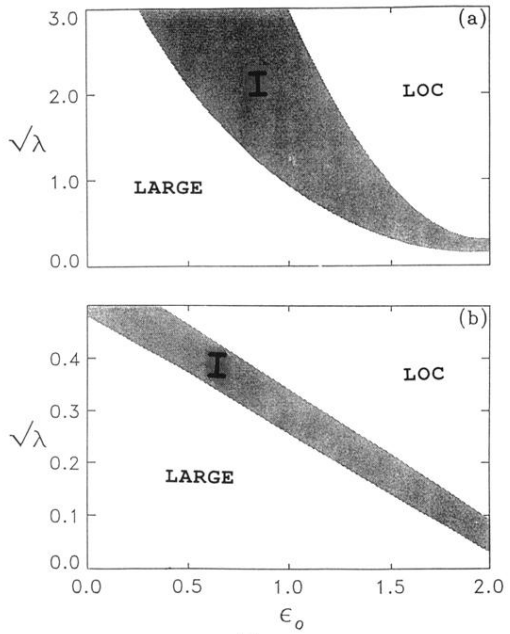

FIG. 3. Separation of ε_0-$\sqrt{\lambda}$ plane into regions of large, intermediate (I), and localized (LOC) polarons for initial energy (a) at the center of the band, $E_e = 0$, and (b) near the bottom of the band, $E_e = -1.949$.