Instanton approach to the lattice polaron problem

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(Received 2 August 1993)

Starting from the path-integral approach to the process of tunneling in the presence of phonons we develop a theoretical procedure for the calculation of the bandwidth of the small-radius polaron in a one-dimensional crystal lattice. A two-site analog of the Holstein molecular crystal model is explored to study the tunneling character in the limit of small phonon frequencies. We do not observe any change-over from the regime of Frank-Condon type of bandwidth renormalization to the so-called adiabatic regime anticipated in the molecular crystal model.

I. INTRODUCTION

An electron interacting with elastic deformations of the host lattice is usually called a polaron. From the theoretical point of view, the study of a single polaron is equivalent to the study of a particle moving in a periodic potential and interacting with a phonon field. In this general sense the problem appears to be relevant to a wide class of phenomena in solid state physics including the physics of excitons,[1,2] the behavior of defects in alkali halides,[3] and light interstitials in metals.[4]

An enormous literature (see, e.g., Refs. 1, 2, 5, and 6) is devoted to the properties of the polaron systems which have now been studied for more than five decades. It is well known that the ideas of polaron theory for Jahn-Teller compounds have played a stimulating role in Bednorz and Müller's experiments[7] crowned by the discovery of high-temperature superconductivity (HTSC). Therefore it is not surprising that the strong-coupling polaron theory is considered by a number of authors[8–13] to be the basis for understanding this new phenomenon.

In the strong-coupling limit as the size of lattice distortion becomes comparable to the lattice constant, one is speaking about a small polaron state as a nearly localized ("self-trapped") state. This means an electron moving together with the surrounding phonon cloud in a narrow polaronic band. The width of the latter is usually expected to be considerably reduced with respect to the bare electron bandwidth by the oscillator wave function overlap factor being known in the literature as Debye-Waller, Frank-Condon, or Lang-Firsov renormalization factor. The value of the polaron bandwidth appears to be a very important parameter for all the properties of small polaron systems, and in particular in the role which they may play in the superconducting state of high-T_c material.

It is commonly accepted that all the essential physics of the small polaron state can be adequately described on the basis of Holstein's molecular crystal model (MCM)[14] containing the minimal set of important parameters for the interacting electron-phonon system. In the simplest version these are the phonon frequency ω_0 of molecular vibrational breathing mode, the polaron binding energy E_p, and the nearest-neighbor tunneling matrix element J. The parameter space of MCM is described by a plot of Fig. 1 which turns out to be helpful in analyzing the different regimes of polaron problem. The phonon renormalization factor is given by the exponential function of the dimensionless interaction constant $g^2=E_p/\omega_0$ which may become large in the small polaron region determined by

$$\frac{E_p}{J} \geq 1 . \tag{1}$$

The latter inequality defines the strong-coupling limit and is known in the polaron literature as the condition for small polaron formation.

As was argued by Holstein,[14] the phonon overlap factor $\exp(g^2)$ governs the bandwidth renormalization in the so-called antiadiabatic regime when the phonon frequency is quite large. In the opposite case of small phonon

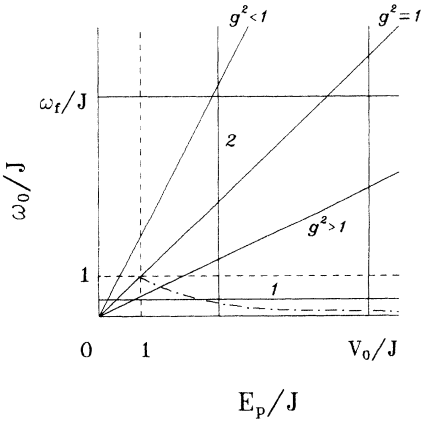

FIG. 1. Two-dimensional parameter space of the molecular crystal model.

frequencies one comes into the adiabatic regime with a different type of expression for the polaron bandwidth. According to Holstein the crossover between the adiabatic and the antiadiabatic regime is given by $J \sim g\omega_0$ (see dashed-dotted line in Fig. 1), this estimate being obtained by equating two different expressions for the bandwidth.

There is also another *ad hoc* prescription for the crossover to occur at $J \sim \omega_0$ (Ref. 2) which seems to be questionable since it does not depend on the interaction strength. For an example of quite a different point of view we refer to Ref. 15 where only the phonon overlap type of renormalization was anticipated for the entire small polaron region given by Eq. (1).

The purpose of the present paper is to discuss the small polaron properties starting from the more theoretically justified description of the tunneling process in the presence of phonons. Such an approach was initiated by Sethna[16] (see also Ref. 17) and may be considered as a generalization of Feynman path-integral formalism to the lattice polaron problem. This description is based on (i) the concept of tunneling as a quasiclassical motion along an (instanton) path in an configurational space and (ii) the field theoretical methods for locating this path for the tunneling entity. The method of integrating out the phonon variables (which is due to Feynman) reduces the problem to that of the motion of a particle in the effective potential with an additional retarded self-interaction term. The main difference from the MCM and most other theoretical approaches is that here the bare tunneling amplitude J is not introduced as a given parameter containing "all" the information about the tunneling in the rigid lattice. It is, on the contrary, a calculable quantity which moreover appears not to be of very crucial importance for the analysis of tunneling process in the presence of phonons. As was pointed out by Sethna, the truncation of a noninteracting particle Hamiltonian to a matrix containing only one parameter J (being a prerequisite of MCM), referred to by him as "truncation approximation," may lead to substantial oversimplification of the model resulting in the loss of its physical significance under certain conditions. We present here evidence that precisely this might happen in the above mentioned "adiabatic" regime of small polaron tunneling.

In the following section of this paper we provide a general description of the path-integral approach to the one-dimensional lattice polaron problem in the zero-temperature limit. An essential part of the calculation dealing with Euclidean action, the most important quantity entering the formula for the polaron bandwidth in the exponential form, is given in Sec. III. We also give an approximate expression for the preexponential factor thus providing the possibility of evaluating the polaron bandwidth explicitly. Section IV contains the qualitative analysis of the two-site polaron model which serves as a good teaching example for any lattice polaron theory. The formalism is developed in close analogy with the molecular crystal model. The question of small polaron crossover from "adiabatic" to "antiadiabatic" regime is of special concern in this section. We cannot find any physical reason for this crossover to take place. Moreover, there is also no indication for the bare polaron bandwidth to be of any significance in the discussion of this crossover.

II. GENERAL DESCRIPTION OF THE INSTANTON APPROACH

We start with the Lagrangian L for a single electron specified by a coordinate x, moving in a one-dimensional periodic potential $V(x)$ of a host lattice of vibrating molecular units located at the minima of $V(x)$: $x = x_n$ (the internal degrees of freedom of these vibrating molecular units are denoted by q_n)

$$L = \tfrac{1}{2}\mu\dot{x}^2 - V(x) + \frac{1}{2}M\sum_n (\dot{q}_n^2 - \omega_0^2 q_n^2) - \lambda\sum_n q_n W(x - x_n) \, . \tag{2}$$

Here μ and M denote the mass of the electron and of the vibrating molecules, respectively, λ is the strength of the electron-phonon interaction, chosen with its usual linear in q dependence; $W(x - x_n)$ describes the electron-lattice interaction as a function of the distance of the electron from the nth molecule and is assumed to have maximal value at $x = x_n$: $W(0) = 1$. In order to have a close analogy with the Holstein model, $W(x - x_n)$ is supposed to vanish for $|x - x_n| > a/2$ (a is the lattice period) although the general formalism does not depend on this assumption.

The statistical density matrix for the system

$$\rho(x, \{q_n\}; x', \{q_n'\}; \beta) = \sum_{m,k} \Psi_{m,k}(x, \{q_n\}) \Psi^*_{m,k}(x', \{q_n'\}) e^{-\beta E_m(k)} \, , \tag{3}$$

where m is the electron band index and k the electron momentum, contains only low-lying energy levels, when considered at low temperature ($\beta \to \infty$). We assume those to have a narrow band structure with the renormalized (polaron) tunneling amplitude $\tilde{J}$ being much smaller than the phonon frequency ω_0,

$$\tilde{J} \ll \omega_0 \, . \tag{4}$$

It should be noted that this inequality, serving as a criterium for validity of the formalism developed here, does not imply the smallness of the bare electron tunneling amplitude J as compared to ω_0.

With the inverse temperature β going to infinity, only the lowest polaron band contribution survives in Eq. (3). Restricting ourselves to this limit we integrate formally over the $\{q_n\}$ variables taking for simplicity $q_n' = q_n = q_{n0}$ and transfer the sum over k into the integral thus arriving at the result

$$\rho(x, x'; \beta)_{\beta \to \infty} \to \int \frac{a\, dk}{2\pi} \varphi_k(x, x') e^{-\beta E_0(k)} \, , \tag{5}$$

where φ_k is a temperature-independent function.

The same quantity on the other hand has the equivalent path-integral representation

$$\rho(x,x';\beta)=\int_x^{x'} Dx(\tau)\int_{-\infty}^{+\infty}\prod_n dq_{n0}\int_{q_n=q_{n0}}^{q_n'=q_{n0}}\prod_n Dq_n(\tau)e^{-S_E}\ , \tag{6}$$

with

$$S_E=\int_{-\beta/2}^{+\beta/2}d\tau\left\{\frac{1}{2}\mu\dot{x}^2(\tau)+V[x(\tau)]+\frac{1}{2}M\sum_n[\dot{q}_n^2(\tau)+\omega_0^2q_n^2(\tau)]+\lambda\sum_n q_n(\tau)W[x(\tau)-x_n]\right\} \tag{7}$$

being the Euclidean action of the system.

It is a textbook result of Feynman[18] that the integration over the phonon trajectories can be carried out explicitly leading to the one-electron problem with a retarded self-interaction term in the effective action:

$$\rho(x,x';\beta)=\rho_{\rm ph}(\beta)\int_x^{x'}Dx(\tau)\exp\{-S_{\rm eff}[x(\tau)]\}\ , \tag{8}$$

$$S_{\rm eff}[x(\tau)]=\int_{-\beta/2}^{+\beta/2}d\tau\{\tfrac{1}{2}\mu\dot{x}^2(\tau)+V[x(\tau)]\}-\sum_n\frac{\lambda^2}{4M\omega_0}\int\int_{-\beta/2}^{+\beta/2}d\tau\,d\tau' G(\tau-\tau')W[x(\tau)-x_n]W[x(\tau')-x_n]\ , \tag{9}$$

$$G(\sigma)=[n_{\rm ph}e^{\omega_0|\sigma|}+(n_{\rm ph}+1)e^{-\omega_0|\sigma|}],\quad n_{\rm ph}=(e^{\beta\omega_0}-1)^{-1}\ . \tag{10}$$

The factorized out bare-phonon matrix density

$$\rho_{\rm ph}(\beta\to\infty)\to\exp\left[-\beta\sum_n\frac{\omega_0}{2}\right] \tag{11}$$

contributes with the oscillator zero-point energy to the ground state energy of the whole system and will be neglected for the following discussion.

Defining the position of the nth molecule to be

$$x_n=a(n-\tfrac{1}{2}),\quad n=0,\pm1,\pm2,\ldots\ , \tag{12}$$

we need to consider $\rho(-a/2,a/2;\beta)$ as the quantity which contains the entire information about the electron tunneling process from one molecular site to another.

A good description of the calculation of the electron bandwidth in the instanton approach without phonons can be found in.[19] The basic entity is the one-instanton tunneling trajectory of the particle, starting at $\tau=-\beta/2\to-\infty$ at $n=0$ site and finishing at $\tau=+\beta/2\to+\infty$ at the adjacent $n=1$ site. When there is no interaction with phonons this trajectory is determined from the classical equation of motion in the inverted potential $-V(x)$:

$$\mu\ddot{x}=V'(x)\ . \tag{13}$$

To make use of the known analytic solution of this equation for the double-well potential

$$V_{d-w}(x)=V_0\left[\frac{4x^2}{a^2}-1\right]^2\ , \tag{14}$$

given by

$$x(\tau)=\frac{a}{2}\tanh\left[\frac{\omega_f}{2}\tau\right],\quad \omega_f=4\sqrt{2V_0/\mu a^2}\ , \tag{15}$$

we fix the host lattice potential $V(x)$ by iterating the tunneling barrier part of $V_{d-w}(x)$ periodically along the x axis (see Fig. 2). The bare tunneling matrix element J which is simply by a factor of 4 smaller than the electron bandwidth is expressed as[19,20]

$$J=\omega_f\sqrt{6S_0/\pi}e^{-S_0}\ , \tag{16}$$

where S_0, the value for the minimum action, corresponding to the instanton path (15), is given by

$$S_0=\frac{16}{3}\frac{V_0}{\omega_f}\ . \tag{17}$$

The quasiclassical description works well for $S_0\geq 6$.[20]

The important physical quantity entering the discussion is ω_f which represents twice the electron ground-state energy in the effective single-well potential of one molecule. According to (15) it defines what Sethna calls the flip time

$$\tau_f=\frac{1}{\omega_f}\ , \tag{18}$$

which it takes the particle to move through the potential barrier between adjacent sites. The increase of τ_f leads to the increase of action due to the potential energy term, while the decrease of τ_f increases action via the kinetic energy term. From the balance of these two contributions one can easily estimate τ_f (Ref. 17) and get the result corresponding to Eq. (15) up to a minor numerical factor.

An instanton calculation of the particle bandwidth implies the condition[20]

$$J\ll\omega_f\ , \tag{19}$$

which means that the flip time has to be much less than

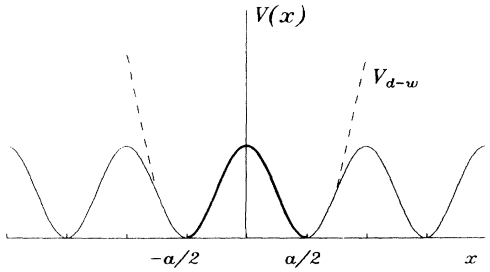

FIG. 2. The periodic host lattice potential of the one-dimensional polaron problem.

the time $\tau_L \sim J^{-1}$ spent by the particle on the site between two subsequent tunneling events. This is known as the dilute instanton gas approximation and insofar as it remains good for the bare electron problem due to the large value of S_0 one expects no difficulty when the interaction with phonons is turned on.

III. ONE-DIMENSIONAL POLARON BANDWIDTH AT ZERO TEMPERATURE

To proceed with the calculation of the tunneling amplitude one has to find the paths for which the action (9) has a local minima. It seems plausible that such a path describing a single tunneling event between the sites $n=0$ and $n=1$ will have the same qualitative behavior as that given by Eq. (15) but with the only important difference in the value of "renormalized" flip frequency $\tilde{\omega}_f$ which replaces the bare flip frequency ω_f. While the precise effect of the retarded interaction term in Eq. (9) can be studied numerically, as it was shown by Sethna[16] there are at least two natural approximations which can be treated on the qualitative level. These are the slow- and fast-flip approximations.

The slow-flip approximation which appears to be good for the case of tunneling of heavy defects[16,17] corresponds to large values of phonon frequency ω_0. Roughly speaking, when ω_0 is the highest frequency of the problem ($\omega_0 \gg \omega_f$), one can replace $G(\sigma)$ in Eq. (9) by $2\delta(\sigma)/\omega_0$ thus arriving at

$$\begin{aligned} S_{\text{eff}}[x(\tau)] &= \int d\tau \{ \tfrac{1}{2}\mu \dot{x}^2(\tau) + U[x(\tau)] - E_p \} \\ &\equiv S_{\text{local}}[x(\tau)] - \beta E_p \ , \\ U(x) &= V(x) - E_p \left[\sum_n W^2(x - x_n) - 1 \right] , \end{aligned} \tag{20}$$

where $U(x)$ is the effective potential experienced by the particle instantaneously followed by the lattice and $E_p = \lambda^2/2M\omega_0^2$ is the previously mentioned polaron shift. The effective potential U retains the translational symmetry of the lattice and if it has no unexpectedly large deviations from V, the resulting flip time will be of the same order as the initial one. A substantial change of the basic trajectory and the flip rate can be imagined for some peculiar forms of W and unreasonably large values of polaron shift E_p, comparable with the ground-state energy of the particle in a single well. We shall not deal with such a situation in this paper.

What is of much greater interest for the polaron problem is the second, fast-flip approximation corresponding to $\omega_0 \ll \omega_f$. Before analyzing this limit it is convenient to transform Eq. (9) for S_{eff} using the relation

$$G(\sigma) = \frac{2}{\omega_0}\delta(\sigma) + \frac{1}{\omega_0^2}\frac{\partial^2}{\partial \sigma^2} G(\sigma) \ .$$

The first term on the right-hand side (rhs), as in the slow-flip case, gives rise to the effective potential $U(x)$, while the second one, after a double partial integration, results in a velocity dependent nonlocal contribution to the action which can now be rewritten as

$$S_{\text{eff}}[x(\tau)] = -\beta E_p + S[x(\tau)] \ , \tag{21}$$

$$\begin{aligned} S[x(\tau)] &= S_{\text{local}}[x(\tau)] \\ &+ \frac{1}{2} g^2 \sum_n \int\int d\tau\, d\tau' G(\tau - \tau') W_x'[x(\tau) - x_n] \\ &\times W_x'[x(\tau') - x_n] \dot{x}(\tau) \dot{x}(\tau') \ , \end{aligned} \tag{22}$$

with S_{local} being the same as in Eq. (20). In the fast-flip limit the main contribution to the nonlocal part of $S[x(\tau)]$ comes from the flip region of the instanton trajectory: $\tau \sim \tau' \leq \omega_f^{-1} \ll \omega_0^{-1}$. To the leading approximation one can replace $G(\sigma)$ by its maximum value $2n_{\text{ph}} + 1 = G(0) \to 1$ ($\beta \to \infty$) thus arriving at the result

$$S[x(\tau)] = S_{\text{local}}[x(\tau)] + g^2 \ . \tag{23}$$

Bearing in mind that the polaron tunneling amplitude $\tilde{J}$ is exponentially dependent on S, one immediately identifies the g^2 term in Eq. (23) with the phonon overlap renormalization factor of the small polaron theory. It is noteworthy that in analogy with the slow-flip case (up to not so much important difference between U and V potentials), there is no considerable change in the instanton trajectory and the corresponding flip rate in the fast-flip regime. This might seem questionable in view of the apparent possibility of having very large values of g^2 upon decreasing the phonon frequency. From a first glance analysis of the nonlocal contribution in Eq. (22) one could expect to get a sizable reduction of this term by causing the electron to have a small velocity, which means making it wait for the lattice motion. This is exactly what one supposes to occur in the adiabatic regime $\omega_0 \to 0$. We shall postpone any further discussion of this question to the next section.

If the classical instanton trajectory $x_{\text{cl}}(\tau)$ which gives the minimum value to the effective action Eqs. (9) or (20) is found, the WKB expression for the path-integral Eq. (8) with ρ_{ph} having dropped out reads

$$\begin{aligned} \rho(-a/2, a/2; \beta) &= e^{\beta E_p} \int_{-a/2}^{+a/2} Dx(\tau) e^{-S[x(\tau)]} \\ &= e^{\beta E_p} N \{ \text{Det} B[x_{\text{cl}}(\tau)] \}^{-1/2} e^{-S[x_{\text{cl}}(\tau)]} \ . \end{aligned} \tag{24}$$

The preexponential factor takes into account the quadratic fluctuations in the vicinity of $x_{\text{cl}}(\tau)$. Apart from the correctly determined normalization constant N it contains the determinant of the nonlocal differential operator

$$B = \frac{\delta^2 S}{\delta x(\tau) \delta x(\tau')} \ , \tag{25}$$

evaluated on the classical instanton trajectory. We estimate the preexponent of (24) by using as a guideline the analogous procedure for the bare tunneling process presented in Ref. 20. At the first step one introduces a somewhat simpler "reference" operator $B_0 = B[x(\tau) = x_m]$ corresponding to the particle motion in the vicinity of the arbitrary minimum of the effective potential $U(x)$. Omitting for the time being the polaron shift factor $\exp(\beta E_p)$ one has

$$\rho(-a/2,a/2;\beta) = N[\mathrm{Det}B_0]^{-1/2}\left\{\frac{\mathrm{Det}B[x_{\mathrm{cl}}(\tau)]}{\mathrm{Det}B_0}\right\}^{-1/2} e^{-S_{\mathrm{cl}}} \quad (26)$$

with S_{cl} being a shorthand notation for the action at the instanton trajectory. An important property of $B[x_{\mathrm{cl}}(\tau)]$ (but not of B_0) is the presence of a zero eigenvalue which makes Eq. (26) formally divergent. This arises from the apparent invariance of the original Lagrangian with respect to time shifts. The shift of the instanton center $\tau\rightarrow(\tau-\tau_c)$ in Eq. (15) cannot result in any change of action. Intuitively it should seem obvious that the instanton must enter Eq. (24) in the form of an integral over the position τ_c of its center. Separating out this part of the path integral[20] is equivalent to the substitution

$$\left\{\frac{\mathrm{Det}B[x_{\mathrm{cl}}(\tau)]}{\mathrm{Det}B_0}\right\}^{-1/2} \rightarrow \left\{\frac{\mathrm{Det}'B[x_{\mathrm{cl}}(\tau)]}{\mathrm{Det}B_0}\right\}^{-1/2} \sqrt{S_{\mathrm{cl}}/2\pi}\,d\tau_c\ , \quad (27)$$

which appears to be of rather general importance in the instanton calculus. In Eq. (27) Det′ denotes the reduced determinant with the zero eigenvalue having been removed. For the simple double well Eq. (14) the curly bracket on the r.h.s. of Eq. (27) is equal to $\sqrt{12}\omega_f$.[20] This result is the same for our choice of $V(x)$ (Fig. 2) when the interaction with phonons is switched off. Assuming the results with interaction turned on to be not too much different from this value, we write down the final expression for the one-instanton tunneling amplitude which included all the classical trajectories in interval $d\tau_c$ near $\tau_c=0$:

$$\rho(-a/2,a/2;\beta)_{\text{one-inst}} = N[\mathrm{Det}B_0]^{-1/2}(\sqrt{6S_{\mathrm{cl}}/\pi}e^{-S_{\mathrm{cl}}}\omega_f)d\tau_c\ . \quad (28)$$

The quantity

$$\tilde{J}=\omega_f\sqrt{6S_{\mathrm{cl}}/\pi}e^{-S_{\mathrm{cl}}} \quad (29)$$

has the meaning of an instanton density in the time space.

Direct integration of Eq. (28) over $d\tau_c$ which is equivalent to replacing $d\tau_c$ by β yields an expression for ρ up to a good approximation as long as $\beta\tilde{J}$ remains much less than unity. As β tends to infinity one has to consider multi-instanton (anti-instanton) paths[19,20] thus arriving at

$$\rho(-a/2,a/2;\beta)=N[\mathrm{Det}B_0]^{-1/2}\sum_{n,\bar{n}}\frac{(\beta\tilde{J})^{n+\bar{n}}}{n!\bar{n}!}\delta_{n-\bar{n},1}\ . \quad (30)$$

Using the identity

$$\delta_{n-\bar{n},1}=\int_{-\pi/a}^{+\pi/a}\frac{a\,dk}{2\pi}e^{ika(n-\bar{n}-1)}\ , \quad (31)$$

one can easily rewrite Eq. (30) as

$$\rho(-a/2,a/2;\beta)=N[\mathrm{Det}B_0]^{-1/2} \times\int_{-\pi/a}^{+\pi/a}\frac{a\,dk}{2\pi}e^{-ika}e^{\beta[E_p+2\tilde{J}\cos(ka)]}\ . \quad (32)$$

In Eq. (32) we have put back the previously omitted polaron shift factor. Comparison of this expression with Eq. (5) provides a strict identification of $\tilde{J}$, Eq. (29), with the renormalized polaron bandwidth.

The only quantity undetermined up to now remains the determinant of the B_0 operator. When the interaction with phonons is turned off this is reduced to the determinant of the harmonic oscillator[20]

$$N[\mathrm{Det}B_0]^{-1/2}=\sqrt{\mu\omega_f/\pi}e^{-\beta\omega_f/2},\quad (\lambda=0,\ \beta\rightarrow\infty) \quad (33)$$

contributing with the ground-state energy of a particle localized in the single potential well of the molecular site. We develop an approximate method of $\mathrm{Det}B_0$ calculation in the Appendix. Here we quote the result which reads

$$N[\mathrm{Det}B_0]^{-1/2}=\sqrt{(\mu/\pi)\omega_1\omega_2/\omega_0}e^{-\beta(\omega_1+\omega_2-\omega_0)/2} \quad (34)$$

with frequency parameters $\omega_{1,2}$ being given by Eq. (A9). After the substitution of this expression into Eq. (32) one can readily obtain the dispersion of the polaron in its lowest energy band

$$E_0(k)=-E_p+\tfrac{1}{2}(\omega_1+\omega_2-\omega_0)-2\tilde{J}\cos(ka)\ . \quad (35)$$

All the quantities entering this formula can be evaluated explicitly. As it was pointed out in the preceding discussion the developed formalism remains valid as far as renormalized bandwidth $\tilde{J}$ remains small compared with the frequency of phonon vibrations and the flip frequency of the tunneling particle.

IV. ON THE EXISTENCE OF ADIABATIC REGIME FOR SMALL POLARON TUNNELING

When discussing polaron properties one is usually concerned with the self-trapping transition between large- and small-radius polaron states. In the framework of the MCM presumably the most natural way to study this transition region is to analyze the behavior of the relevant quantities along the horizontal line 1 in Fig. 1, that is to calculate them as a function of the effective coupling E_p/J for a fixed value of the adiabaticity parameter ω_0/J (see, e.g., Ref. 15). Extensive studies in this direction were carried out in the context of the two-site polaron problem[21–27] which represents the simplest version of the MCM, containing all the important features of polaron states.

Less attention was paid to the changeover of polaron properties as a function of phonon frequency when the strength of the interaction with phonons remains fixed. As mentioned in the Introduction, it is generally believed that the so-called adiabatic regime exists for sufficiently small values of ω_0. However, no concensus has emerged up to now clearly defining the boundary of this regime

within the relevant parameter space. It is the purpose of this paper to discuss this question starting from the path-integral approach to the two-site polaron problem. In this case there is only one phonon mode (of the two) which couples to the electron motion between the two molecular sites.[21,27] This enables us to write down the corresponding Lagrangian function as follows:

$$L=\tfrac{1}{2}\mu\dot{x}^2-V_{d-w}(x)+\tfrac{1}{2}M(\dot{q}^2-\omega_0^2q^2)-\frac{\lambda}{\sqrt{2}}q\Phi(x)\ , \tag{36}$$

where

$$\Phi(x)=-\Phi(-x)\equiv W(x-x_0)-W(x-x_1)$$

describes the effective interaction of electron with the out-of-phase $q=(q_0-q_1)/\sqrt{2}$ vibrational mode involving the $n=0$ and $n=1$ sites.

To retain the analogy with the Holstein model as much as possible, $\Phi(x)$ should be taken as approximately constant (±1) in the vicinity of the sites ($x=\pm a/2$) and display some transitory behavior near $x\simeq0$. A smooth curve 1 depicted in Fig. 3(a) may be quite adequate in representing all the necessary features of $\Phi(x)$. Curve 2 with ν as a free parameter will be used in our further analysis as a convenient explicit parametrization.

We intend to consider the small polaron limit which is approached by the condition (1) in the Holstein model and corresponds to a strong reduction of the tunneling amplitude ($\tilde{J}<\omega_0$) either by the phonon overlap exponent or via some other renormalization law in the—presumably existing—adiabatic regime.

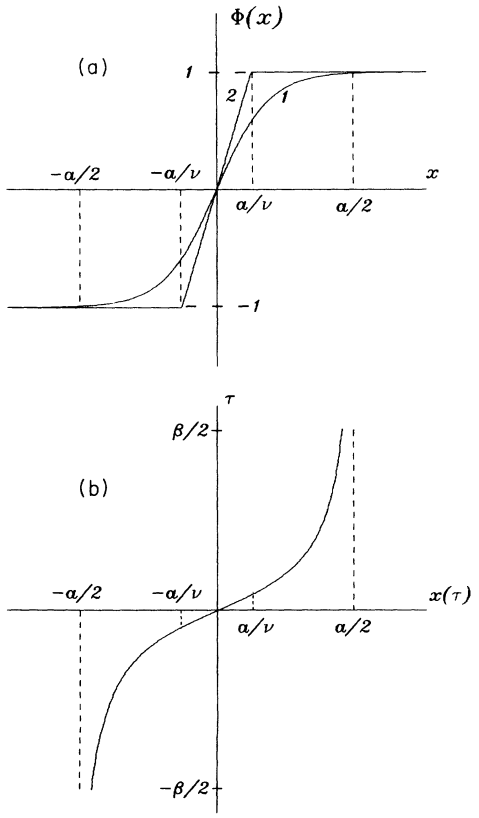

FIG. 3. (a) The electron-phonon interaction intensity as a function of the electron coordinate in the two-site polaron problem. (b) The classical (instanton) trajectory describing a single tunneling event.

The expression for the Euclidean action which is strictly analogous to our previous expression Eqs. (21) and (22) reads

$$S[x(\tau)]=S_{\text{local}}[x(\tau)]+S_{\text{nonlocal}}[x(\tau)]\ , \tag{37}$$

$$S_{\text{local}}[x(\tau)]=\int_{-\infty}^{+\infty}d\tau(\tfrac{1}{2}\mu\dot{x}^2(\tau)+V_{d-w}[x(\tau)]-\tfrac{1}{2}E_p\{\Phi^2[x(\tau)]-1\})\ , \tag{38}$$

$$S_{\text{nonlocal}}[x(\tau)]=g^2\frac{1}{4}\int\int_{-\infty}^{+\infty}d\tau\,d\tau'\Phi_x'[x(\tau)]\Phi_x'[x(\tau')]\times\dot{x}(\tau)\dot{x}(\tau')e^{-\omega_0|\tau-\tau'|}\ , \tag{39}$$

where the zero-temperature limit is used from the very beginning in as far as the quantities under consideration must remain finite when β tends to infinity. The polaron shift $E_p=g^2\omega_0$—being independent on M—is taken to be fixed as well as all other parameters which enter Eq. (38) for the local part of the Euclidean action. By decreasing the phonon frequency, that is by going down along the vertical line 2 in Fig. 1, one gets an unlimited increase of g^2 which multiplies the nonlocal contribution to the action, Eq. (39). We need to verify whether or not the corresponding growth of S_{nonlocal} may be compensated by an appropriate decrease in the particle velocity $\dot{x}(\tau)$ which is controlled by the flip-time parameter of the instanton trajectory. If this happens one can think of the electron as waiting for the lattice to move in the process of transition across the barrier.

We assume the classical trajectory, giving the minimum value for the effective action Eqs. (37)–(39), to have the same qualitative time dependence as that of Eq. (15). The flip frequency has possibly a different value $\tilde{\omega}_f=\xi\omega_f$ with ξ being the frequency renormalization parameter. We proceed further by parametrizing $\Phi(x)$ according to Fig. 3(a) (curve 2) as

$$\Phi(x)=\begin{cases}\nu x/a & \text{if } |x|\le a/\nu\\ \pm1 & \text{otherwise}\ ,\end{cases} \tag{40}$$

where ν is supposed to be large (say not less than 4). As a result only a small region of the instanton trajectory being essentially linear in τ,

$$x(\tau)\simeq\frac{a}{4}\tilde{\omega}_f\tau\ , \tag{41}$$

contributes to the nonlocal part of the action [see Fig. 3(b)].

From Eqs. (39)–(41) one easily gets

$$S_{\text{nonlocal}}/g^2\equiv I=\frac{1}{4}\int_{-1}^{+1}dy\int_{-1}^{+1}dy'e^{-\alpha|y-y'|}\ , \tag{42}$$

where

$$\alpha=\frac{\omega_0}{\tilde{\omega}_f}\frac{4}{\nu}=\frac{\Delta}{\xi},\quad \Delta=\frac{\omega_0}{\omega_f}\frac{4}{\nu}\ . \tag{43}$$

The integral on the rhs of Eq. (42) has an upper bound equal to unity corresponding to $\alpha=0$ and decays as $1/\alpha$ for large α. For our estimates we find it convenient to parametrize Eq. (42) in the simplest approximate form $I=1/(1+\alpha)$ which is expected to work well in both the small and, most importantly, the large α regions.

For the reasonable values of E_p and the proposed form of $\Phi(x)$ one can hardly expect the effective potential contribution arising from the electron-phonon interaction to play an important role in S_{local}. We therefore shall ignore in the following the term in Eq. (38) proportional to E_p. Thus, the essential part of S_{local} is related to the bare potential and the kinetic energy terms. For the instanton path Eq. (15) both of them contribute equally to S_0 [Eq. (17)]. The rescaling of the flip frequency ($\omega_f\rightarrow\xi\omega_f$) can be easily treated by a corresponding rescaling of the time variable leading to $S_{\text{local}}=S_0(\xi+\xi^{-1})/2$.

As a result we have the following expression for the effective action of the two-site problem:

$$S[x(\tau)]=\frac{1}{2}S_0\left[\xi+\frac{1}{\xi}\right]+g^2\frac{\xi}{\xi+\Delta}\ . \tag{44}$$

The value of ξ which minimizes S is determined by the equation

$$\frac{1}{\xi^2}=1+\frac{2g^2\Delta}{S_0}\frac{1}{(\xi+\Delta)^2}\ . \tag{45}$$

One readily finds that the solution to Eq. (45) obeys the inequalities

$$\sqrt{1-2g^2\Delta/S_0}\le\xi\le 1 \tag{46}$$

with the lower boundary being determined by the ratio

$$\frac{2g^2\Delta}{S_0}=\frac{2E_p}{\omega_f S_0}\frac{4}{\nu}=\frac{3}{8}\frac{E_p}{V_0}\frac{4}{\nu}\ll 1\ . \tag{47}$$

This appears to be remarkably independent of ω_0 and is inevitably small as long as $E_p\ll V_0$.

We thus come to the conclusion that the flip frequency remains essentially unchanged when ω_0 tends to zero, providing the fast-flip approximation and the $\exp(g^2)$ type of bandwidth renormalization works well in this limit. We find no indication for an adiabatic regime within this path-integral approach for the local type of electron-phonon interaction which characterizes the MCM. There is, moreover, no indication for the bare electron bandwidth to be of primary importance for our discussion of tunneling properties.

V. CONCLUSIONS

Starting from the path-integral approach to the quantum tunneling process in the periodic potential we have developed a theoretical formalism describing the band motion of a particle in the presence of phonons. By applying this to the lattice polaron problem we have proposed a method for direct evaluation of polaron bandwidth in the small polaron regime of the one-dimensional analog of Holstein's molecular crystal model. The initial (bare) tunneling amplitude appears to be of minor significance in extracting the important physical features of the system, which are primarily determined by the value of the flip-time parameter being absent in the canonical picture of polaron motion. We definitely do not observe any changeover of tunneling properties when entering the region of adiabaticity however small ω_0 might be. The fast-flip (Frank-Condon) approximation with a bandwidth controlled by the phonon overlap is found to cover the entire parameter space which generally is considered to be governed by the adiabatic approximation. For the local type of electron-phonon interaction one can see no "driving force" which could make the electron slow down its across-barrier transition in adjusting to the motion of the lattice.

ACKNOWLEDGMENTS

The authors would like to acknowledge valuable discussions of the various aspects of the problem with A. Alexandrov, V. Kabanov, and J. Ranninger. One of the authors (A.K.) expresses his gratitude to the director of CRTBT-CNRS, P. Monceau, for his hospitality during the author's stay in Grenoble.

APPENDIX

We present here the expressions for the differential operators B and B_0 and propose a procedure for the calculation of $\mathrm{Det}B_0$.

The operator B, which is defined by Eq. (25), reads

$$B=\delta(\tau-\tau')\left\{-\mu\frac{\partial^2}{\partial\tau^2}+U''[x(\tau)]+2E_p\sum_n\{W'[x(\tau)-x_n]\}^2\right\}$$
$$+\delta(\tau-\tau')\sum_n W''[x(\tau)-x_n]F_n(\tau)-\omega_0E_pG(\tau-\tau')\sum_n W'[x(\tau)-x_n]W'[x(\tau')-x_n]\ , \tag{A1}$$

where

$$F_n(\tau)=W[x(\tau)-x_n]-\tfrac{1}{2}\omega_0\int d\bar{\tau}\,G(\tau-\bar{\tau})W[x(\bar{\tau})-x_n]\ .$$

It is reduced to the operator B_0

$$B_0=\delta(\tau-\tau')\left\{-\mu\frac{\partial^2}{\partial\tau^2}+U''(x_m)+2E_p\sum_n(W'[x_m-x_n])^2\right\}-\omega_0E_pG(\tau-\tau')\sum_n(W'[x_m-x_n])^2\,, \tag{A2}$$

for $x(\tau)=x_m, x_m$ being any arbitrary minimum of the effective periodic potential $U(x)$.

Defining the quantities

$$\mu R=E_p\sum_n[W'(x_m-x_n)]^2\,,\qquad \mu\Omega^2=U''(x_m)+2\mu R\,, \tag{A3}$$

we rewrite the expression for B_0 in the more compact form

$$B_0=\delta(\tau-\tau')\left[-\mu\frac{\partial^2}{\partial\tau^2}+\mu\Omega^2\right]-\mu\omega_0RG(\tau-\tau')\,. \tag{A4}$$

We proceed further by defining an auxiliary operator Q corresponding to a harmonic oscillator of mass μ and frequency ω_0 whose determinant is a well-known quantity

$$Q=\left[-\mu\frac{\partial^2}{\partial\tau^2}+\mu\omega_0^2\right],\quad N^{-2}\mathrm{Det}Q=\frac{\pi}{\mu\omega_0}e^{\beta\omega_0}\,. \tag{A5}$$

One can obviously express $\mathrm{Det}B_0$ as

$$\mathrm{Det}B_0=\mathrm{Det}(QB_0Q^{-1})=\mathrm{Det}(QB_0)/\mathrm{Det}Q\,. \tag{A6}$$

After evaluating the kernel of the product operator QB_0

$$\begin{aligned}K(QB_0)&=\int d\tau''\delta(\tau-\tau'')Q(\tau'')B_0(\tau''-\tau')\\&=\left[-\mu\frac{\partial^2}{\partial\tau^2}+\mu\omega_0^2\right]\left[\delta(\tau-\tau')\left[-\mu\frac{\partial^2}{\partial\tau'^2}+\mu\Omega^2\right]-\mu\omega_0RG(\tau-\tau')\right]\\&=\delta(\tau-\tau')\left[\left[-\mu\frac{\partial^2}{\partial\tau'^2}+\mu\omega_0^2\right]\left[-\mu\frac{\partial^2}{\partial\tau'^2}+\mu\Omega^2\right]-2\mu^2\omega_0^2R\right],\end{aligned} \tag{A7}$$

we can write

$$\begin{aligned}QB_0&=\left[-\mu\frac{\partial^2}{\partial\tau^2}+\mu\omega_0^2\right]\left[-\mu\frac{\partial^2}{\partial\tau^2}+\mu\Omega^2\right]-2\mu^2\omega_0^2R\\&=\left[-\mu\frac{\partial^2}{\partial\tau^2}+\mu\omega_1^2\right]\left[-\mu\frac{\partial^2}{\partial\tau^2}+\mu\omega_2^2\right],\end{aligned} \tag{A8}$$

where

$$\omega_{1,2}^2=\tfrac{1}{2}[(\omega_0^2+\Omega^2)\pm\sqrt{(\omega_0^2-\Omega^2)^2+8\omega_0^2R}\,]\,. \tag{A9}$$

We are now able to express $\mathrm{Det}B_0$ in terms of simple oscillator determinants

$$\mathrm{Det}B_0=\mathrm{Det}\left[-\mu\frac{\partial^2}{\partial\tau^2}+\mu\omega_1^2\right]\times\mathrm{Det}\left[-\mu\frac{\partial^2}{\partial\tau^2}+\mu\omega_2^2\right]\Big/\mathrm{Det}Q\,, \tag{A10}$$

thus arriving at the final result

$$N^{-2}\mathrm{Det}B_0=\frac{\pi}{\mu}\frac{\omega_0}{\omega_1\omega_2}e^{\beta(\omega_1+\omega_2-\omega_0)}\,. \tag{A11}$$

The combination of frequencies $(\omega_1+\omega_2-\omega_0)$ in the exponent may be interpreted as twice the ground-state energy of the particle when it is measured from the bottom of the effective potential well. One can easily check that this quantity tends to the value of ω_f if the interaction with phonons is switched off.

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