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PHYSICAL REVIEW B VOLUME 7, NUMBER 10 15 MAY 1973

Ground State of the Exciton-Phonon System

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(Received 25 October 1972)

The ground state of a model exciton-phonon Hamiltonian is studied using variational techniques. A single-exciton band is considered in the tight-binding model; the exciton is coupled to Einstein phonons through a short-range linear interaction. We first verify that a variational wave function corresponding to simple displacements of the lattice coordinates (analogous to the Lee–Low–Pines wave function for the polaron) leads to an unphysical result: For strong exciton-phonon coupling the effective mass of the excitons depends discontinuously on the parameters of the Hamiltonian. We obtain an improved trial function by studying an exactly soluble problem: an exciton hopping between two sites and coupled to a phonon field. The new trial function allows distortion of the Gaussian form of the phonon wave function as well as displacement. Analogous trial functions are used to calculate the energy and effective mass for a one-dimensional lattice with nearest-neighbor exciton hopping. The results are a continuous effective mass and a substantial improvement of the ground-state energy over the Lee–Low–Pines trial function. Arguments are given that the qualitative behavior of the ground state is independent of the dimensionality of the lattice, so that the one-dimensional calculation performed here is adequate.

I. INTRODUCTION

The interaction of an exciton or an electron with lattice vibrations leads to a number of interesting effects.[1–3] If the exciton-phonon interaction is weak, the major effects are a reduction in mobility due to scattering, the introduction of phonon sidebands in optical absorption,[4–6] a small change in mass, etc. For very strong coupling the phenomenon of self-trapping[7,8] occurs. Here, the distortion of the lattice in the vicinity of the exciton leads to a large increase in effective mass. At high temperatures, the motion of the particle through the lattice changes from band type to activated hopping.[9]

This paper is concerned with the transition be-

tween nearly free and self-trapped behavior as the exciton-phonon coupling strength is increased. Ever since Landau[7] first introduced the idea of self-trapping, there has been speculation as to whether the onset of this phenomenon occurs discontinuously as a function of the coupling strength, or whether there is a smooth transition between the two types of behavior. In order to discuss this question, we must specify the nature of the particle-phonon interaction carefully. When the particle is an electron the usual interaction is of the Fröhlich-polaron type,[10] in which the electron interacts with the polarization field of the lattice via the long-range Coulomb interaction. The transition from small to large coupling of the Fröhlich polaron has been extensively studied,[11] and it appears that the polaron mass is a continuous function of the coupling parameter, i.e., that self-trapping does not have a sudden onset. However, in a variational calculation for a Fröhlich polaron bound by a weak Coulomb potential, Larsen[12] has shown that a sharp transition may occur, so even in this case the question cannot be regarded as finally settled.

When the particle is an exciton, the interaction is usually short range. As a model, we can regard the exciton as exerting a force only on those atoms in the same unit cell of the lattice as its own instantaneous location. This model may also be valid for the electron-phonon interaction in nonpolar crystals, where the interaction could occur via the deformation potential.[13] The character of the transition between weak and strong coupling has been much less clear in this case. Toyozawa[13] has pointed out that the absence of a sudden onset of Landau trapping of polarons may be due to the long range of the electron-lattice interaction. Using a Lee–Low–Pines[14] form of wave function, he has given a variational calculation in a case where the interaction is short range (interaction of an electron with acoustic phonons); in that calculation self-trapping manifests itself as a discontinuity of the effective mass of the ground state as a function of coupling strength. A further conclusion of that variational calculation is that for intermediate values of coupling the free and self-trapped states can coexist; as the coupling increases the energies of the two states cross, with the self-trapped state having lower energy for strong coupling. A similar calculation has been done for molecular excitons by Merrifield.[15] The lack of discontinuities in the numerical calculation of that paper is apparently due to the fact that the exciton-phonon coupling strength was always too weak.

Experimental evidence of a discontinuous mass in exciton or electron systems is unclear. Fivaz and Mooser[16] have interpreted experiments on the layerlike semiconductors MoS_2 and GaSe in terms of a model similar to Toyozawa's which contains discontinuities. No actual jumps seem evident in their data, however.

More recently, Cho and Toyozawa[17] have studied the optical absorbtion of the exciton-phonon system nonvariationally, by diagonalizing the Hamiltonian within a restricted subspace of phonon states. In contrast to the earlier result, they find no discontinuity in effective mass or other properties. Instead, they find a dependence of effective mass on coupling constant which is "smooth, but quite abrupt." In related work, Sumi[18] has shown that large (unbound) and small (bound) polarons can coexist for certain values of the coupling parameter. For different values of coupling strength, one type or the other can have lower energy, but the character of the transition is still not clear.

In the following sections we seek to clarify the situation, discuss the inadequacy of the Lee–Low–Pines wave function for this problem, and introduce improved variational wave functions that produce a lower ground-state energy and a continuous mass in the intermediate region. We will argue that these functions give a better representation of the transition to the self-trapping regime than the Lee–Low–Pines function. In Sec. II we use a Tamm–Dancoff method to investigate the role that the dimensionality and coupling strength play in the self-trapping problem. We conclude that there is no essential difference in the behavior of one or three dimensional systems. In Sec. III we study variational approximations to a problem that can be solved numerically: an exciton hopping between two sites and coupled to a phonon field. We develop an improved variational wave function that can be extended to the N-site problem. In Sec. IV we calculate the ground-state energy and effective mass for the N-site problem using improved variational functions. In Sec. V we discuss the physical significance of the new variational wave functions and conclusions concerning the character of the ground state.

The Hamiltonian for the interacting exciton-phonon system is written as

$$\mathcal{H} = -\frac{\Delta_0}{2z}\sum_{\sigma} c_{i+\sigma}^{\dagger} c_i + \omega \sum_i a_i^{\dagger} a_i - \frac{\omega (W_0)^{1/2}}{2} \sum_i c_i^{\dagger} c_i (a_i + a_i^{\dagger}) \quad . \tag{1}$$

This Hamiltonian describes a linear, simple square, or simple cubic lattice. The first term describes the free exciton band, of width Δ_0. Here z is the number of nearest neighbors, equal to twice the dimensionality of the system; the index σ runs over nearest neighbors. The operator $a_i^{\dagger} a_i$ is the number operator for the harmonic oscillator

at lattice site i; all oscillators have frequency ω. The last term describes the exciton-phonon interaction, which is assumed to be local and linear in oscillator displacement. The strength of the interaction is given by the dimensionless parameter W_0. The binding energy of the exciton in the phonon field for zero bandwidth ($\Delta_0 = 0$) is $-\omega W_0/4$. The Hamiltonian $\mathcal{H}$ is identical to that of Ref. 17 with the following replacements: $\omega \rightarrow 1$, $\Delta_0 \rightarrow 2B$, $W_0 \rightarrow 4S$. This model has several limitations. Since only a single exciton band is considered, there is no possibility of an exciton-phonon bound state due to internal motion of the exciton, as considered by Toyozawa and Hermanson.[19] Further, this model yields an infinite mean free path at $T = 0$, since the phonons are dispersionless. Still, this model seems appropriate for investigating variational approximations to the ground state, and particularly the onset of self-trapping.

II. NATURE OF SELF-TRAPPING POTENTIALS

In this section we argue that the dimensionality of the system does not play a crucial role in the self-trapping problem. This contention is in apparent contradiction to the well-known fact that a weak, attractive, short-range static potential always leads to a bound state in one dimension, but does not in three dimensions.[20] We show that a feature of three-dimensional static potentials, namely, that they must exceed a certain strength to lead to binding is absent in self-trapping. The difference between the self-trapping and static-potential problems can be made clear by the following calculation.

For an electron interacting with an attractive static potential V_0, we can write a model Hamiltonian:

$$\mathcal{H}_1 = -\frac{\Delta_0}{2z}\sum_{i\sigma} c_{i+\sigma}^\dagger c_i - V_0 \sum_i c_i^\dagger c_i a_i^\dagger a_i \quad . \tag{2}$$

Here, the operator $a_i^\dagger a_i$ refers to a fictitious particle at site i that attracts the electron with potential $-V_0$. We first assume this particle is located at the origin and look for an eigenfunction of the form

$$\Psi_1 = \sum_j g_j c_j^\dagger a_0^\dagger |0\rangle \quad , \tag{3}$$

where the g_j are constants to be determined.

It is very easy to show that the eigenvalues E_1 of $\mathcal{H}_1$ are solutions of the equation

$$\frac{V_0}{N}\sum_q \frac{1}{\epsilon_q - E_1} = 1 \quad , \tag{4a}$$

where

$$\epsilon_q = -\frac{\Delta_0}{2z}\sum_\sigma e^{i\vec{q}\cdot\vec{\sigma}} \quad . \tag{4b}$$

The eigenfunctions are then given by

$$g_j = \frac{V_0}{N} g_0 \sum_q \frac{e^{i\vec{q}\cdot\vec{R}_j}}{\epsilon_q - E_1} \quad . \tag{4c}$$

We now ask whether Eqs. (4) possess a bound-state solution $E_1 < -\frac{1}{2}\Delta_0$. If they do, the electron is "trapped," since the g_j will then fall off exponentially for large R_j. As is well known, the answer depends on the dimensionality. In one dimension, the sum $\sum_q (\epsilon_q + \frac{1}{2}\Delta_0)^{-1}$ diverges for $|q| \to 0$, so that a bound state must exist. For three dimensions, the extra q^2 factor in the numerator produces a finite sum, leading to a condition on V_0 for the existence of a bound state:

$$\frac{V_0}{N}\sum_q (\epsilon_q + \tfrac{1}{2}\Delta_0)^{-1} > 1 \quad . \tag{5}$$

In order to obtain a translationally invariant eigenstate of $\mathcal{H}_1$, analogous to the $k = 0$ state of an exciton, Ψ_1 can be replaced by

$$\Psi_1' = \frac{1}{\sqrt{N}}\sum_{j\delta} g_\delta c_{j+\delta}^\dagger a_j^\dagger |0\rangle \quad . \tag{6}$$

This eigenstate has exactly the same properties as Ψ_1, since there is no coupling between terms of different j.

We now wish to follow the argument of Eqs. (2)–(6) as closely as possible for the case of the self-trapped exciton. We modify Eq. (1) slightly to emphasize the similarity with Eq. (2):

$$\mathcal{H}_2 = -\frac{\Delta_0}{2z}\sum_{i\sigma} c_{i+\sigma}^\dagger c_i + \omega \sum_i a_i^\dagger a_i - (\tfrac{1}{2}V_0\omega)^{1/2}\sum_i c_i^\dagger c_i (a_i + a_i^\dagger) \quad . \tag{7}$$

The usual argument for self-trapping consists of looking for an "adiabatic" solution to Eq. (7), in which the exciton is assumed to move in a "static" potential produced by the distortion of the lattice. The over-all translational invariance is then restored by summing a linear combination of such adiabatic terms.

A simple version of the adiabatic argument consists of ignoring the hopping term in Eq. (7). Then, the adiabatic potential is obtained by assuming the exciton is fixed at the origin, $\langle c_i^\dagger c_i\rangle = \delta_{i,0}$, and diagonalizing the phonon terms. The result, $\langle a_i + a_i^\dagger\rangle = (2V_0/\omega)^{1/2}\delta_{i,0}$, leads to an adiabatic potential $V_i^{\mathrm{ad}} = -V_0\delta_{i,0}$. If one then argues that this potential can only exist if V_0 is large enough to bind the exciton at the origin in the first place, then the analogy with the static potential seems complete and the conclusion follows that in three dimensions a small V_0 will not lead to self-trapping.

The simple argument can be improved, without changing the conclusion, by requiring that the adiabatic potential be self-consistent. If the ex-

citon is bound, with a wave function

$$\Psi=\sum_i g_i c_i^\dagger|0\rangle \ , \tag{7}$$

then we require $\langle a_i+a_i^\dagger\rangle=(2V_0/\omega)^{1/2}|g_i|^2$. This replaces the δ-function potential of the simple argument with a finite but short-range self-consistent potential. The conclusion that there is no bound state in three dimensions unless V_0 exceeds a critical value is still valid.

We will argue here that for small V_0 the adiabatic argument is incorrect since it is improper to solve the phonon problem first and restore the larger hopping term later. Instead, we start with a $k=0$ eigenfunction of the form

$$\Psi_2=\left[\sum_i c_i^\dagger+\sum_{i\delta} a_i^\dagger c_{i+\delta}^\dagger f_\delta\right]|0\rangle \ . \tag{8}$$

For $V_0\ll\omega$, it is correct to limit Ψ_2 to terms containing zero and one phonons. The coefficients f_δ and the eigenvalues are determined by using a Tamm–Dankoff method[10,21]:

$$P(\mathcal{H}_2\Psi_2)=E_2\Psi_2 \ . \tag{9}$$

Here P is a projection operator that projects out of $\mathcal{H}_2\Psi_2$ those parts containing zero or one phonons. The result is

$$E_2=-\frac{\Delta_0}{2}-\frac{V_0\omega}{2N}\sum_q\frac{1}{\omega+\epsilon_q-E_2} \ , \tag{10}$$

$$f_\delta=(\tfrac{1}{2}V_0\omega)^{1/2}N^{-1}\sum_q\frac{e^{i\vec{q}\cdot\vec{R}_\delta}}{\omega+\epsilon_q-E_2} \ . \tag{11}$$

Equation (10) always has a bound-state solution $E_2<-\frac{1}{2}\Delta_0$, whatever the dimensionality or coupling strength. This solution is essentially the second-order perturbation energy, obtained by replacing E_2 by $-\frac{1}{2}\Delta_0$ in the denominator of the q summation. Further, this denominator never vanishes, so that the qualitative features of the solution do not depend on the dimensionality of the summation. The coefficients f_δ fall off exponentially with distance even for a weak potential in three dimensions.

These conclusions are not an approximation, but are rigorous features of the model. This becomes clear if one considers that the above calculation is, in fact, variational. Since we have found a bound state below the bottom of the band, the exact ground state must have a still lower energy. We conclude that there are no essential features of the self-trapping problem that depend on the dimensionality of the model. Further, in raising the question of a sudden onset of self-trapping as the interaction energy V_0 is increased, the preceding example gives convincing evidence that this onset could not occur as a transition to a "localized" state in which the dynamic lattice potential finally becomes large enough to bind the electron or exciton. There remains the possibility that a discontinuity could occur in a manner suggested in Ref. 13, where the effective strength of the lattice potential itself, i.e., the distortion of the lattice produced by the exciton, varies discontinuously as the interaction parameter is changed. In the remainder of this paper we shall concentrate on this latter possibility.

III. TWO-SITE PROBLEM

The argument of the preceding section leads to the speculation that the spatial form of the wave function will always be exponential and may not be of critical importance in studying the transition between untrapped and trapped behavior of the exciton. This is in contrast to the case of the static attractive potential, where the change from power law to exponential form of the wave function defines the onset of the bound state. We expect then that we can gain useful information about the transition by studying a simpler problem that can be solved exactly with a computer: an excitation hopping between two sites. The intention is to use the computer solution for the wave function to construct a variational wave function that is an improvement upon functions of the type used by Toyozawa[13] or Merrifield[15] and then generalize the improved wave function for the many-site problem.

The Hamiltonian is[22]

$$\mathcal{H}=-\Delta_0 S_x-\omega(W_0)^{1/2}S_z(a+a^\dagger)+\omega a^\dagger a \ . \tag{12}$$

The two states are represented by a spin-$\frac{1}{2}$ particle; S_x, S_z are spin operators. The "tunneling" parameter Δ_0 governs the motion between the sites (flipping the spin). As in Eq. (1), W_0 is a dimensionless measure of the coupling strength. The sign of the coupling of the "spin" to the single harmonic oscillator depends on whether the spin points up or down.

For small Δ_0 the ground state of $\mathcal{H}$ is

$$\Psi=2^{-1/2}\left[\begin{pmatrix}1\\0\end{pmatrix}e^{-(W_0)^{1/2}(a-a^\dagger)/2}+\begin{pmatrix}0\\1\end{pmatrix}e^{+(W_0)^{1/2}(a-a^\dagger)/2}\right]|0\rangle \ . \tag{13}$$

Here $|0\rangle$ is the phonon vacuum. If $u=W_0\omega/\Delta_0\gg1$, Eq. (13) gives a good approximation to the ground state. In Eq. (13) the spin points in a given direction long enough for the oscillator to relax completely. The interpretation in terms of a displaced harmonic oscillator is made obvious by defining a dimensionless position operator:

$$x=(1/\sqrt{2})(a+a^\dagger) \ . \tag{14}$$

In the x representation, the wave function (13) becomes

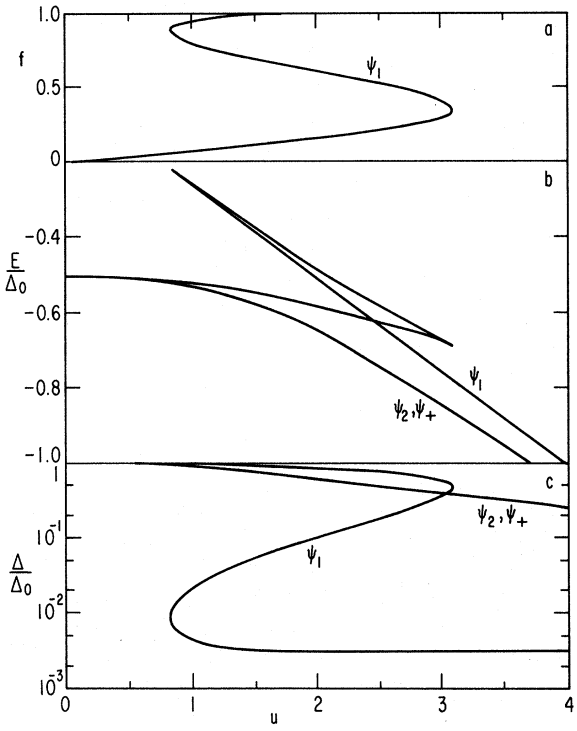

FIG. 1. Properties of the two-site model for $W_0=12$. (a) Values of f that produce extremal values of $\langle\mathcal{H}\rangle$ in Eq. (17), vs the parameter $u=\omega W_0/\Delta_0$. (b) Energy obtained from the variational functions Ψ_1 and Ψ_2 and the exact solution Ψ_+. The energies obtained from Ψ_2 and Ψ_+ are indistinguishable on the scale of this plot. (c) Renormalized tunneling parameter Δ vs u, calculated using several trial functions. The curve for Ψ_2, Ψ_+ asymptotically approaches the curve for Ψ_1 at very large values of u. The value of Δ calculated from Ψ_1 changes discontinuously at the critical value of u, $u_c\approx 2.4$.

$$\Psi(x)=2^{-1/2}\left[\begin{pmatrix}1\\0\end{pmatrix}\Psi_+(x)+\begin{pmatrix}0\\1\end{pmatrix}\Psi_-(x)\right], \tag{15a}$$

where

$$\Psi_\pm(x)=\pi^{-1/4}\exp\{-\tfrac{1}{2}[x\mp(\tfrac{1}{2}W_0)^{1/2}]^2\}. \tag{15b}$$

In the opposite extreme $u\ll 1$, the ground state is found by replacing W_0 by 0 in Eq. (13). In this limit the spin-flip rate Δ_0 is so large that the oscillator always remains in its undisplaced ground state. To find a solution for intermediate values of u, an obvious first choice is to treat the displacement as a variational parameter. This is analogous to the variational solution used in Ref. 13. We put

$$\Psi_1=\frac{1}{\sqrt{2}}\,e^{-fS_z(W_0)^{1/2}(a-a^\dagger)}\begin{pmatrix}1\\1\end{pmatrix}|0\rangle\ . \tag{16}$$

Here f is a variational parameter which is expected to vary from 0 to 1 as u increases. The expectation value of the energy is

$$\langle\Psi_1|\mathcal{H}|\Psi_1\rangle=-\tfrac{1}{2}\Delta_0 e^{-f^2W_0/2}+\tfrac{1}{4}\omega W_0(f^2-2f)\ . \tag{17}$$

This expression is minimized with respect to f. For fixed W_0, we find that the equation $\partial\langle\mathcal{H}\rangle/\partial f=0$ has three roots for a certain range of values of u in the vicinity of $u=1$, provided that W_0 is sufficiently large ($W_0>\frac{27}{4}$). Two of the roots correspond to local minima of $\langle\mathcal{H}\rangle$ and one root to a local maximum. These results are illustrated in Figs. 1(a) and (1b) for $W_0=12$, where the ground-state energy and f are plotted as a function of u.

For u smaller than a critical value u_c, the absolute minimum of $\langle\mathcal{H}\rangle$ corresponds to the smaller of the two choices of f (typically $f\approx 0$). As u increases through u_c, f shifts discontinuously to the larger extremal value ($f\approx 1$). This result can be related to self-trapping by defining a "renormalized" tunneling parameter $\Delta\equiv\langle 2\Delta_0 S_x\rangle$. We associate Δ with the "bandwidth" of the exciton; the inverse of Δ gives a measure of the "effective mass." If we use Ψ_1 to calculate Δ,

$$\Delta=\Delta_0 e^{-f^2W_0/2}\ . \tag{18}$$

As u increases through u_c the discontinuity in f leads to a sudden exponential decrease in Δ, as shown in Fig. 1(c). This discontinuity is a primitive form of self-trapping discussed in Ref. 13.

Further, in the region $u\sim u_c$, the two minima in $\langle\mathcal{H}\rangle$ can be interpreted as describing a ground and excited state with greatly different Δ. For $u\lesssim u_c$ the ground state is untrapped and the excited state is trapped. For $u\gtrsim u_c$ the reverse is true. This feature of Ψ_1 is also discussed in Ref. 13.

It is clear that the wave function Ψ_1 gives an adequate description of the ground state for $u\gg 1$ and $u\ll 1$. However, we now show that the discontinuous onset of self-trapping at $u=u_c$ is an artifact of the simple form of the wave function Ψ_1; in reality Δ is a continuous function of u. This can be established, along with information needed to construct an improved variational wave function, by obtaining the exact ground state of Eq. (12) numerically. The numerical solution is facilitated by performing a unitary transformation on $\mathcal{H}$:

$$\mathcal{H}'=U\mathcal{H}U^{-1}, \tag{19a}$$

where

$$U=2^{-1/2}\begin{pmatrix}1 & R\\ -1 & R\end{pmatrix}\ , \tag{19b}$$

$$R=(-1)^{a^\dagger a}\ . \tag{19c}$$

The operator R obeys

$$R=R^{-1}\ ,\quad R^\dagger=R\ ,\qquad [R,a]_+=[R,a^\dagger]_+=0\ . \tag{20}$$

The result is

$$\mathcal{H}'=-\Delta_0 S_z R-\tfrac{1}{2}\omega(W_0)^{1/2}(a+a^\dagger)+\omega a^\dagger a\ . \tag{21}$$

$\mathcal{H}'$ is diagonal in the spin coordinate; the eigen-

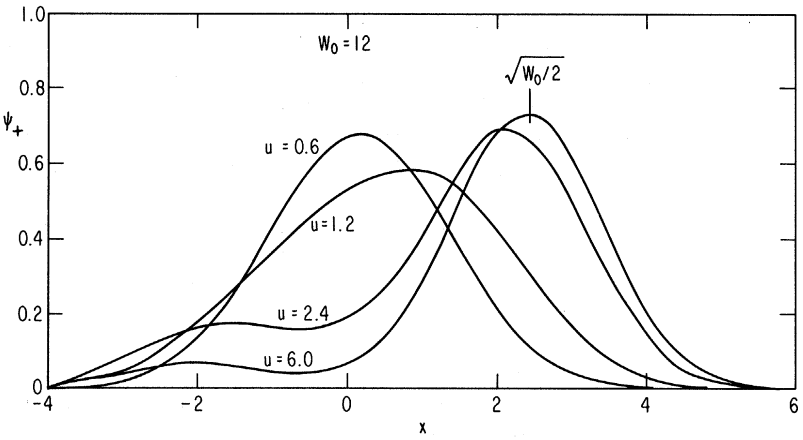

FIG. 2. The wave function Ψ_+ calculated from Eq. (23) vs x for the two-site model. Several values of u are shown for fixed W_0.

states separate into two manifolds, obtained by setting $S_z = \pm\frac{1}{2}$. These correspond, respectively, to the symmetric and antisymmetric combinations of the original spin-up and spin-down states. The two states are analogous to an exciton with $k = 0$ and k at the zone boundary.

The ground state is obtained by setting $S_z = \frac{1}{2}$ and numerically diagonalizing a matrix $H'_{m,n}$ with nonzero elements:

$$H'_{n,n} = -\tfrac{1}{2}\Delta_0(-1)^n + \omega n \ ,$$
$$H'_{n,n+1} = H'_{n+1,n} = -\tfrac{1}{2}\omega(W_0)^{1/2}(n+1)^{1/2},$$
$$n = 0,\ 1,\ \ldots \ . \qquad (22)$$

The result for the ground-state energy is shown in Fig. 1(b) for $W_0 = 12$. The parameter Δ is shown in Fig. 1(c). Since the energy, eigenvector, and Δ are continuous functions of u, we conclude that the discontinuity obtained from Ψ_1 is an artifact. The next step is to investigate the ground-state eigenvector to understand the inadequacy of Ψ_1.

Any eigenstate of $\mathcal{H}$ can be written in the form (15a), with exact $\Psi_\pm(x)$ to replace Eq. (15b); the eigenstate of $\mathcal{H}'$ corresponds to Ψ_+. In the x representation,

$$\Psi_+(x) = \pi^{-1/4} e^{-x^2/2} \sum_{n=0}^{\infty} \alpha_n (2^n n!)^{-1/2} H_n(x) \ , \qquad (23)$$

where the α_n are the coefficients of the ground-state eigenvector of $\mathcal{H}'$ and the $H_n(x)$ are Hermite polynomials of order n. In Fig. 2 we plot $\Psi_+(x)$ for $W_0 = 12$ and several values of u. From the figure it is obvious that Ψ_+ cannot be represented by a single displaced harmonic oscillator for values of u in the transition region $u \sim u_c$. This is apparently the source of failure of Ψ_1. If we calculate the mean displacement $\bar{x}$ of the oscillator using the Ψ_+, we find a continuous variation between $\bar{x} \approx 0$ for $u \ll u_c$ to $\bar{x} \approx (W_0/2)^{1/2}$ for $u \gg u_c$. However, for $u \approx u_c$ the rms zero-point motion $\langle (x - \bar{x})^2 \rangle^{1/2}$ increases above the value $2^{-1/2}$ characteristic of a harmonic oscillator. This behavior is characteristic of all the transitions between trapped and untrapped behavior that we have studied.

We now write down a trial wave function that more closely approximates the behavior described above and which can be generalized to the many-site problem, where a numerical solution cannot easily be obtained. We try a linear combination of two displaced oscillators:

$$\Psi_2 \propto [e^{-(W_0)^{1/2}(a-a^\dagger)/2} + \gamma e^{-g(W_0)^{1/2}(a-a^\dagger)/2}] | 0 \rangle \ . \qquad (24)$$

Here f, g, and γ are variational parameters chosen to minimize the expectation value of $\langle \Psi_2 | \mathcal{H}' | \Psi_2 \rangle / \langle \Psi_2 | \Psi_2 \rangle$. The x-representation wave function $\Psi_2(x)$ is a sum of two Gaussians; this form is clearly suggested by the numerical solution in Fig. 2. The ground-state energy calculated from Ψ_2 is shown in Fig. 1(b) for $W_0 = 12$. On the scale of Fig. 1(b) it is indistinguishable from the exact numerical value. There is no discontinuity in any of the variational parameters. The renormalized tunneling parameter is plotted in Fig. 1(c). Once more the agreement with the numerical solution is extremely good. Thus, there seems to be reasonable justification for modifying the Lee–Low–Pines type of trial function in a manner analogous to Eq. (24).

IV. THE N-SITE PROBLEM

We now attempt to treat an exciton in the infinite lattice, described by the Hamiltonian of Eq. (1). The general solution can be written

$$\Psi_k = \frac{1}{\sqrt{N}} \sum_i e^{i\vec{k}\cdot\vec{R}_i} c_i^\dagger \phi_k(\{Q_{i,\delta}\}) | 0 \rangle \ . \qquad (25)$$

Here k is a wave vector, R_i is the location of the lattice site i, and ϕ is a function of all the oscillator coordinates $Q_{i,\delta}$ (where the oscillator i, δ is located at $\vec{R}_i + \vec{R}_\delta$). From the argument of Sec. II, we know that the term in Ψ_k containing the electron at site i will only affect oscillators in the vicinity of that site. The important features of ϕ_k

are (1) its spatial extent, i.e., the distance over which the lattice oscillators are distorted; and (2) the form of the disturbance for oscillators in the immediate vicinity of the exciton. For fixed W_0, both properties depend on the parameter $u = W_0\omega/\Delta_0$. For $u \gg 1$, all oscillators are in their ground state, except for the oscillator at the site of the electron ($\delta = 0$). The wave function for the $\delta = 0$ oscillator corresponds to a simple displacement by the full value $\langle x_{\delta=0}\rangle = (\frac{1}{2}W_0)^{1/2}$. For $u \ll 1$, the displacement of the $\delta = 0$ oscillator is small $\langle x_{\delta=0}\rangle \approx (\frac{1}{2}u)^{1/2}$, but the disturbance falls off with distance as a slow exponential, involving roughly $(W_0/u)^{1/2}$ neighbors. We will show that for intermediate values of u, the wave function for the $\delta = 0$ oscillator approximates a double Gaussian, as in the two-site problem. The transition from small to large u is again continuous.

Since the qualitative behavior of ϕ, including the exponential character of the dependence on distance, does not depend on the dimensionality of the lattice, we will concentrate on a one-dimensional model, $z = 2$ in Eq. (1). Then, the location of oscillator R_δ is $R_\delta = a\delta$; here a is the nearest-neighbor distance and δ takes on values 0, ± 1, ± 2, ... The ground-state energy and effective mass of the exciton can be identified by expanding the energy for small k:

$$E_k = E_0 + \frac{1}{2m^*}k^2 + \cdots \quad . \tag{26a}$$

For $W_0 = 0$, the effective mass is simply

$$m_0 = 4/\Delta_0 a^2 \quad . \tag{26b}$$

Then, for fixed W_0, the ratio m^*/m_0 varies between 1 and $e^{W_0/4}$ as u increases.

We first display the apparent discontinuity in the effective mass for $u \approx u_c$, using a wave function similar to that of Ref. 13;

$$\Psi_k^I = \frac{1}{\sqrt{N}}\sum_i e^{i\vec{k}\cdot\vec{R}_i} c_i^\dagger \exp\{\tfrac{1}{2}(W_0)^{1/2} \times \sum_\delta (f_\delta^* a_{i+\delta}^\dagger - f_\delta a_{i+\delta})\}\,|\,0\rangle \quad . \tag{27a}$$

The invariance of a momentum eigenstate under the combined operations of spatial inversion and time reversal requires that

$$f_{-\delta} = f_\delta^*, \quad \delta = 0, \pm 1, \pm 2, \ldots \quad . \tag{27b}$$

To calculate E_0, we set $k = 0$; then we can require that all f_δ be real. The expectation value of the energy is

$$\langle \Psi_0^I |\, \mathcal{H} \,| \Psi_0^I\rangle = -\tfrac{1}{2}\Delta_0 \exp\left\{-\tfrac{1}{4}W_0 \sum_{\delta=0}^{\infty} (f_{\delta+1} - f_\delta)^2\right\} + \tfrac{1}{4}\omega W_0(f_0^2 - 2f_0) + \tfrac{1}{2}\omega W_0 \sum_{\delta=1}^{\infty} f_\delta^2 \quad . \tag{28}$$

This has a minimum when the f_δ's are related to $f_{\delta=0}$ by

$$f_\delta = f_0 e^{-\kappa|\delta|} \quad , \tag{29a}$$

where

$$\kappa = \ln\left(\frac{1-f_0}{1+f_0}\right) \quad . \tag{29b}$$

That is, the f's (which measure the distortion of the δth nearest neighbor of the site containing the electron) fall off exponentially with distance.

Using the results of Eqs. (29) in Eq. (28), the expectation value of the energy $\langle \mathcal{H} \rangle$ can be written in terms of f_0 alone:

$$\langle \mathcal{H} \rangle = -\tfrac{1}{2}\Delta_0 e^{-f_0^3 W_0/4} + \tfrac{1}{8}\omega W_0(f_0^3 - 3f_0) \quad . \tag{30a}$$

The structure of this equation is similar to Eq. (17) for the two-site model. When we minimize $\langle \mathcal{H} \rangle$ with respect to f_0, $\langle \mathcal{H} \rangle$ has extrema when

$$\frac{f_0^2}{1-f_0^2}\, e^{-W_0 f_0^3/4} = \frac{\omega}{\Delta_0} \quad . \tag{30b}$$

For large W_0 ($W_0 > 5^5/2\times 3^4 \approx 19.3$), Eq. (30b) has multiple roots, resulting in a discontinuity in f_0 as u increases. The results for the ground-state energy are shown in Fig. 3 for $W_0 = 24$. As in the two-site problem, the second minimum of $\langle \mathcal{H} \rangle$ can be interpreted as an excited state with greatly different effective mass than the ground state.

From Eq. (29) it can be seen that the disturbance is well localized for the trapped state ($f_0 \approx 1$) and is spread out for the untrapped state ($f_0 \approx 0$).

The effective mass for this model is found using Eq. (27) for nonzero k. The calculation is com-

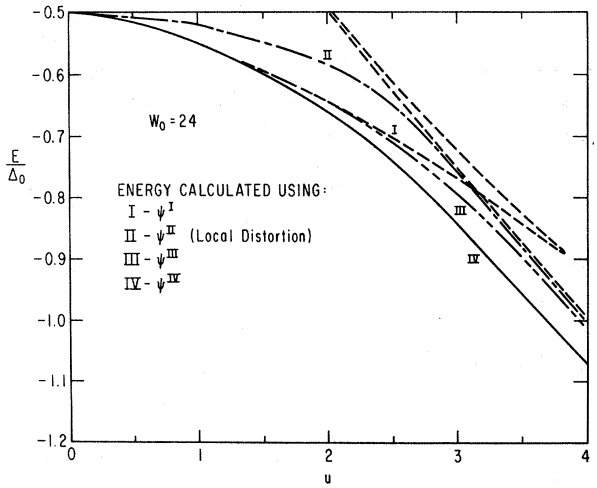

FIG. 3. Expectation value of the energy for the one-dimensional N-site model vs the parameter $u = \omega W_0/\Delta_0$. The trial functions are obtained from I – Eq. (27); II – Diagonalization of the matrix in Eq. (36); III – Eq. (37); IV – Eq. (38).

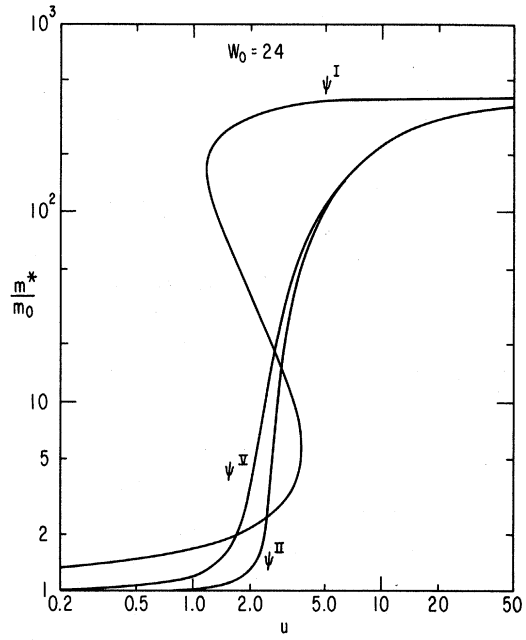

FIG. 4. Calculated effective mass vs u for several trial functions. The mass calculated with Ψ_1 changes discontinuously at the critical value of u, $u_c \approx 3.2$.

plicated by the fact that the f_δ's should be taken to be complex. The expectation value of the energy is

$$\langle \Psi_k^I | \mathcal{H} | \Psi_k^I \rangle = -\tfrac{1}{4}\Delta_0 \exp\Big[iak - \tfrac{1}{4} W_0 \sum_{\delta=0}^{\infty} (|f_\delta|^2 + |f_{\delta+1}|^2 - 2f_\delta^* f_{\delta+1})\Big] + \text{c.c.} + \tfrac{1}{4}\omega W_0 (f_0^2 - 2f_0) + \tfrac{1}{2}\omega W_0 \sum_{\delta=1}^{\infty} |f_\delta|^2 \quad . \tag{31}$$

Calculating $\langle \mathcal{H} \rangle$ to second order in k, it is found that f_0 ($=f_0^*$) is given by Eqs. (30), $f_\delta' = \mathrm{Re} f_\delta$ is given by Eqs. (29), and

$$f_\delta'' = \mathrm{Im} f_\delta = -\frac{f_0 \sinh\kappa}{\sinh\kappa + \frac{1}{2} W_0 f_0^2}\, \delta e^{-\kappa\delta} \quad . \tag{32}$$

Then, letting λ denote $e^{-\kappa}$, the effective mass becomes

$$\frac{m}{m^*} = \frac{\omega}{\Delta_0}\,[1 + \tfrac{1}{8} W_0 f_0 (1 - f_0^2)]^{-2} \times \left\{ 2W_0 f_0^2 S_1 + \frac{1 - f_0^2}{f_0^2}\,(1 + \tfrac{1}{2} W_0 f_0^2 S_2) \right\} , \tag{33a}$$

where

$$S_1 = \sum_{\delta=1}^{\infty} \delta^2 e^{-2\kappa\delta} = \lambda^2(1+\lambda^2)(1-\lambda^2)^{-3} \quad , \tag{33b}$$

$$S_2 = \sum_{\delta=0}^{\infty} [\delta - \lambda(\delta+1)]^2 e^{-2\kappa\delta} = 2\lambda^2(1-\lambda)^2(1-\lambda^2)^{-3} \quad . \tag{33c}$$

The effective mass for $W_0 = 24$ is plotted in Fig. 4.

The discontinuity in f_0 arising from Eqs. (30) is clearly the result of attempting to approximate the wave function for the $\delta = 0$ oscillator by a simple displacement. Another "variational" wave function can be obtained by requiring that all oscillators with $\delta \neq 0$ remain in the ground state; the best wave function for the $\delta = 0$ oscillator can be obtained numerically. This solution is analogous to the "internal subspace" solution of Cho and Toyozawa.[17] We look for an eigenfunction of Eq. (1) of the form

$$\Psi_k^{II} = \frac{1}{\sqrt{N}} \sum_i e^{i\vec{k}\cdot\vec{R}_i}\, c_i^\dagger \phi_k(Q_i) |0\rangle \quad . \tag{34}$$

It is easy to see that, within the subspace of states given by Eq. (34), the minimum energy is obtained when ϕ_k is an eigenfunction of the local Hamiltonian:

$$\mathcal{H}_L^k = \omega a^\dagger a - \tfrac{1}{2}\omega (W_0)^{1/2}(a + a^\dagger) - \tfrac{1}{2}\Delta_0 \cos ak\, |0\rangle\langle 0| \quad . \tag{35}$$

$\mathcal{H}_L^k$ operates on a space spanned by the eigenstates $|n\rangle$ of a single harmonic oscillator:

$$|n\rangle = (n!)^{-1/2} (a^\dagger)^n |0\rangle \quad .$$

The operator $|0\rangle\langle 0|$ is the projection operator for the $n = 0$ state of this oscillator. The ground-state energy of $\mathcal{H}_L^k$ is found by numerically diagonalizing the matrix $(H_L^k)_{mn}$ with nonzero elements:

$$(H_L^k)_{nn} = n\omega - (\tfrac{1}{2}\Delta_0 \cos ak)\delta_{n0} \quad , \qquad (H_L^k)_{n,n+1} = (H_L^k)_{n+1,n} = -\tfrac{1}{2}\omega (W_0)^{1/2}(n+1)^{1/2} \quad . \tag{36}$$

The ground-state ($k = 0$) energy and the effective mass are shown for $W_0 = 24$ in Figs. 3 and 4, respectively. There are no discontinuities in the effective mass, but the restriction to a localized distortion raises the energy considerably above the value obtained using Ψ^I. The approximation is particularly bad for small u, where many neighbors of the electron are in fact displaced. The diagonalization of $(H_L^k)_{mn}$ allows us to determine ϕ_k in the x representation; this is displayed in Fig. 5. The double-Gaussian behavior with large fluctuations of the displacement around its mean value again appears for intermediate values of u.

The preceding results indicate that an improved variational wave function must allow for some spatial extent of the lattice distortion around the exciton position and for a possible "double-Gaussian" form of the distortion. A simple example for a ground-state ($k = 0$) wave function is

$$\Psi^{III} = \frac{1}{\sqrt{N}} \sum_i c_i^\dagger \Big[\exp\Big(-\tfrac{1}{2}(W_0)^{1/2} \sum_\delta f_\delta (a_{i+\delta} - a_{i+\delta}^\dagger) \Big)$$

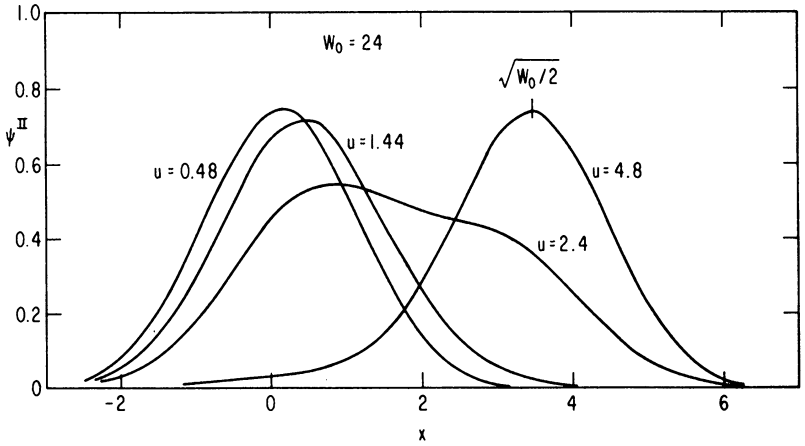

FIG. 5. The x representation for the wave function Ψ^{II} vs x for the one-dimensional N-site model. Several values of u are shown for fixed W_0.

$$+\gamma \exp\left(-\tfrac{1}{2}(W_0)^{1/2}\sum_\delta g_\delta(a_{i+\delta}-a_{i+\delta}^\dagger)\right)\Big]\,|0\rangle \quad . \quad (37)$$

Here the f_δ, g_δ, and γ are (real) variational parameters, obeying $f_\delta = f_{-\delta}$, $g_\delta = g_{-\delta}$; A is a normalization constant. The ground-state energy is shown in Fig. 3 and values of f_δ, g_δ and γ are given in Table I. These values were obtained by minimizing the expectation value of the energy using a standard numerical algorithm and are of uncertain accuracy, since the absolute minimum of a function of a large number of parameters can be very shallow in the multidimensional space of those parameters. We have made sure, however, that the minimum itself is obtained accurately. All parameters are continuous as u is varied and there is a considerable lowering of the energy for all values of u from the value obtained from Ψ^{I}.

The energy can be further lowered for large u by one further change in Ψ^{III}. The ground-state energy asymptotically approaches the value $E \to -\frac{1}{4}\omega W_0$ for large u, corresponding to the energy of a localized displaced harmonic oscillator. The energy can be lowered by order $-\Delta_0^2/\omega W_0$ below this by allowing the exciton to make virtual transitions to nearest-neighbor (undisplaced) oscillators. This results in a wave function in which the lattice displacements are not symmetric with respect to the site of the exciton. The requirement of Ψ^{III} that $f_\delta = f_{-\delta}$, $g_\delta = g_{-\delta}$ inhibits this contribution to the energy. A more suitable trial function is

$$\Psi^{IV} = \frac{A}{\sqrt{N}}\sum_i c_i^\dagger\Big[\exp\left(-\tfrac{1}{2}(W_0)^{1/2}\sum_\delta f_\delta(a_{i+\delta}-a_{i+\delta}^\dagger)\right) + \tfrac{1}{2}\gamma\exp\left(-\tfrac{1}{2}(W_0)^{1/2}\sum_\delta g_\delta(a_{i+\delta}-a_{i+\delta}^\dagger)\right) + \tfrac{1}{2}\gamma\exp\left(-\tfrac{1}{2}(W_0)^{1/2}\sum_\delta g_{-\delta}(a_{i+\delta}-a_{i+\delta}^\dagger)\right)\Big]\,|0\rangle\,. \quad (38)$$

Here $f_\delta = f_{-\delta}$ but there is no restriction on g_δ. The ground-state energy is shown in Fig. 3 and the values of f_δ, g_δ and γ are given in Table II.

Calculations using several trial functions more complicated than Ψ^{IV} do not seem to change the ground-state energy significantly. Consequently we believe that the ground-state energy calculated using Ψ^{IV} is close to the actual value.

TABLE I. Variational parameters to minimize the expectation value of the energy using the trial wave function Ψ^{III}. Here $u = \omega W_0/\Delta_0$; $W_0 = 24$.

	u					
	0.48	1.20	1.92	2.88	3.60	4.56
γ	0.002	0.24	0.54	0.96	2.76	8.59
f_0	0.14	0.19	0.21	0.28	0.29	0.19
$f_{\pm1}$	0.10	0.15	0.19	0.21	0.24	0.37
$f_{\pm2}$	0.08	0.09	0.09	0.09	0.07	0.03
$f_{\pm3}$	0.06	0.06	0.05	0.04	0.02	0.007
g_0	1.0	0.41	0.50	0.79	0.93	0.96
$g_{\pm1}$	•••	0.08	0.08	0.04	0.02	0.008
$g_{\pm2}$	•••	0.09	0.08	0.04	0.02	0.01
$g_{\pm3}$	•••	0.05	0.04	0.02	0.005	0

TABLE II. Variational parameters to minimize the expectation value of the energy using the trial function Ψ^{IV}. $W_0 = 24$.

	u					
	0.48	1.20	1.92	2.88	3.60	4.56
γ	1.89	1.68	1.20	0.46	0.34	0.26
f_0	0.22	0.40	0.57	0.87	0.93	0.96
$f_{\pm1}$	0.13	0.19	0.15	0.05	0.03	0.02
$f_{\pm2}$	0.04	0.04	0.03	0.006	0.002	0.001
g_{-2}	−0.01	−0.002	0.01	0.005	0.002	0.001
g_{-1}	−0.003	0.01	0.02	0.006	0.003	0.002
g_0	0.08	0.17	0.20	0.15	0.13	0.11
g_1	0.13	0.28	0.40	0.77	0.83	0.86
g_2	0.13	0.20	0.19	0.06	0.03	0.02
g_3	0.08	0.10	0.07	0.006	0.002	0.001

We are unable to easily calculate the effective mass using Ψ^{IV}. In order to determine the effect of neighboring lattice sites on m^*, we use the following simpler trial function:

$$\Psi^{V} = \frac{A}{\sqrt{N}} \sum_i e^{i\vec{k}\cdot\vec{R}_i} c_i^\dagger [e^{(W_0)^{1/2} f(a_i^\dagger - a_i)/2} + \gamma e^{(W_0)^{1/2} g(a_i^\dagger - a_i)/2} + \tfrac{1}{2}\beta e^{(W_0)^{1/2}(h^* a_{i+1}^\dagger - h a_{i+1})/2} + \tfrac{1}{2}\beta^* e^{(W_0)^{1/2}(h a_{i-1}^\dagger - h^* a_{i-1})/2}] |0\rangle \quad . \quad (39)$$

The ground-state energy for Ψ^{V} is somewhat higher than that of Ψ^{IV}, particularly for small u where the restriction of the distortion to nearest neighbors is least valid. However, the qualitative features of the solution are similar to Ψ^{IV}. The mass calculated with Ψ^{V} is shown in Fig. 4. As expected, the major effect of including displacements of nearest neighbors is to increase the mass for small u.

V. CONCLUSIONS

The Hamiltonians (1) and (12) describe a fairly wide range of problems in which a spin or fermion interacts linearly with a boson field. Our results indicate that the simple physical idea that these systems can be described by harmonic oscillator wave functions whose mean displacement depends on the hopping rate is not valid if the hopping rate ($\sim\Delta_0$) is comparable to the self energy of the oscillators ($\sim\omega W_0$). In this intermediate regime the fluctuations of the oscillators become anomalously large: the low-lying states of the system are best described as superpositions of nearly completely distorted and completely undistorted oscillators. The fluctuations remove any sign of discontinuities in the effective mass of the excitation, or any sudden onset of self-trapping.

This conclusion is based on particularly firm grounds for the two-site model, where, as we have seen, an exact solution can be given. For the N-site model (i.e., an exciton in a lattice) we could, in general, only give plausibility arguments for our conclusions. In the particular case of Ψ^{II} where distortions are allowed only at the site where the electron resides (i.e., the internal subspace of Ref. 17) we can again give an exact solution. This solution, the two-site model, and the large gains in variational energy from allowing large fluctuations in oscillator wave functions give us confidence in our conclusions.

Similar wave functions to ours have been suggested in quite a different context by Eagles.[23] In his work, attention is focused on the transition between large and small polarons, and he points out that superpositions of the two sorts of states should be considered. This is very close to our contention. It is interesting that in contrast to the case of the static potential, there is little difference in the character of the ground state of a particle coupled to a lattice via a Coulomb potential (the polaron) and the short-range coupling we use here. It was one of the major points of Ref. 13 for example, that major effects could be expected from the difference between long- and short-range potentials. We believe that the discussion of Sec. II shows that the dynamic potential due to the lattice always produces a bound state regardless of the form of the interaction.

*Supported in part by the National Science Foundation.

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