Dimensional mismatch of the electron-phonon system and large polaron stability

Zoran Ivić*
The "Vinča" Institute of Nuclear Sciences, Laboratory for Theoretical and Condensed Matter Physics—020, P.O. Box 522, 11001 Belgrade, Serbia and Montenegro

David W. Brown
University of California, San Diego, CMRR Building, 9500 Gilman Drive, La Jolla, California 92093-0402, USA

Marijana Kirćan
Institut für Theorie der Kondensierten Materie, Universität Karlsruhe, Postfach 6980, 76128 Karlsruhe, Germany
(Received 21 February 2005; revised manuscript received 20 May 2005; published 30 September 2005)

We consider the scaling arguments in the adiabatic limit for materials in which one-dimensional (1D) electronic behavior is influenced by a higher-dimensional vibrational system. The system under consideration consists of a collection of widely separated parallel molecular chains, perfectly one dimensional in respect to their electronic characteristics, embedded in a three-dimensional (3D) lattice. We found that dimensional mismatch between electronic subsystem and lattice may seriously affect polaron stability. For the realistic systems with anisotropic phonon spectrum large polaron stability requires that polaron radius cannot exceed some critical value which is of the order of the interchain spacing. Finally, large polaron stability criterion is formulated in dependence of the values of the basic system parameters.

DOI: 10.1103/PhysRevB.72.094306 PACS number(s): 71.38.−k

I. INTRODUCTION

Investigation of the phenomenon of self-trapping, or polaron formation, has attracted renewed interest recently due to some recent investigations which indicate the existence of polaron carriers in strongly correlated electron-phonon materials such as high-T_c superconductors,[1] colossal magnetoresistance manganites,[2] and organic and quasi-one-dimensional materials.[3,4]

One-dimensional (1D) systems with short-ranged electron-phonon interaction play a special role in polaron theory, since it proves possible to show, on the basis of quite general theoretical arguments[5–7] that, in the adiabatic limit (electron bandwidth large with respect to the maximal phonon energy), they support the existence of stable, mobile, pulse shaped solitonlike large polaron states. Of particular and historical importance in this regard are the scaling arguments of Emin and Holstein,[6] which clearly show that this is not the case in two and three dimensions, which under similar conditions appear to be dominated by states of infinite radius (free states) or states of negligible radius (self-trapped states).

The possibility of realizing solitonlike excitations in one-dimensional systems has naturally generated widespread interest in quasi-1D electron (exciton-) phonon systems.[8–14] It was mainly motivated by the assumed relevance of the polaron (soliton) mechanism in the charge (electron, hole,…) and energy (intramolecular vibrational energy) transfer in quasi-1D conductors (some organic salts and conjugated polymers) and molecular chains such as α helix, acetanilide (ACN) and DNA macromolecules.[3,4,14] Most of the studies on the subject were based upon the idealized 1D models. Such an approach has been very successful in the study of polarons and solitons in conjugated polymers. However, some time ago it was argued that the inevitable small interchain coupling in conducting polymers is large enough to destabilize polaron.[8,9] In particular, Emin[9] established, on the basis of the calculations within the Holstein molecular crystal model, the large polaron stability criterion, which requires at least a third order-of-magnitude anisotropy of transfer integrals. However, according to available data,[15] the ratio of intrachain to interchain transfer integrals is estimated to be of the order of 10 to 10^2 only. These values are considerably lower than that required by Emin's criterion, which means that the acceptance of those estimates would lead to a conclusion that the large polaron cannot be formed in those quasi-1D solids. Nevertheless, experiments including the infrared absorption,[16] charge carrier mobility measurements[17] and resonant Raman spectra[18] of conjugated polymers strongly support polaron presence in these materials. Mizes and Conwell[19] showed that the existence of chain endings and other conjugation breaks can stabilize polarons. Furthermore, Gogolin[20] pointed out that the small-polaron effect causes an additional anisotropy of electron bands so that Emin's criterion may be satisfied. For these reasons, electronic subsystem, in the first approximation at least, may be treated as perfectly one dimensional. On the contrary, the above arguments are not valid for the vibrational subsystem. Namely, in the realistic substances in general, we encounter highly anisotropic electronic subsystems embedded in 3D lattice. The vibrational characteristics of such system cannot be perfectly one dimensional, however, and so we address the dimensional mismatch which must exist between the electronic and vibrational characteristics of any "quasi-one-dimensional" material. In this paper we wish to broaden the scope of the Emin-Holstein scaling arguments in order to consider what effect, if any, three-dimensional character of the host solid may have on polaron properties in 1D chains imbedded in it; that is, we examine the solids built up from molecular chains that are perfectly one dimensional in their electronic characteristics.

We shall use the scaling arguments in the adiabatic limit and regard all real space variables as continuous coordinates. To review the salient features of previous works, we note that the adiabatic theory, based upon the applying the variational method[5] in combination with scaling approach,[6–9,21] leads to the construction of the adiabatic energy functional (Hamiltonian) from which the main features of the self-trapped states may be determined.

II. MODEL HAMILTONIAN AND VARIATIONAL METHOD

We adopt the usual approach and base our analysis on a Fröhlich Hamiltonian of the form

$$H = -J\sum_n B_n^\dagger (B_{n+1} + B_{n-1}) + \sum_{\vec{q}s} \hbar\omega_{qs} a_{\vec{q}s}^\dagger a_{\vec{q}s} + \frac{1}{\sqrt{N}} \sum_{n,\vec{q}s} F_{\vec{q}s} e^{i\vec{q}\cdot\vec{e}_x nR_0} B_n^\dagger B_n (a_{\vec{q}s} + a_{-\vec{q}s}^\dagger). \quad (1)$$

For a 1D electronic system interacting through the short-range forces with the vibrational system of possibly higher dimensionality, the electron-phonon coupling parameter is given by[22] $F_{\vec{q}s} = 2i\chi(\hbar/2M\omega_{qs})^{1/2}(\vec{e}_{\vec{q}s}\cdot\vec{e}_x)\sin(\vec{q}\cdot\vec{e}_x R_0)$ in which $\vec{e}_{\vec{q}s}$ denote phonon modes polarization vectors, $\vec{e}_x$ is the unit vector in the direction of the "chain," which we take to be the "x" axis. Remaining labels are as usual; operator $B_n^\dagger$ (B_n) describes the presence (absence) of the excess electron on the nth site of molecular chain, J is the nearest-neighbor intrachain electronic transfer integral, interchain hopping is neglected, $a_{\vec{q}s}^\dagger$ and $a_{\vec{q}s}$ are creation and annihilation operators of lattice modes $(\omega_{qs};\vec{q},s)$, χ denotes the strength of electron-phonon interaction, and finally R_0 is the lattice constant along the chain. We shall consider the case of an electron coupling with longitudinal acoustic phonons for which polarization vector is simply $\vec{e}_{\vec{q}} = \vec{q}/q (q = |\vec{q}|)$ and therefore the index s will be omitted hereafter. In purely 1D systems $\vec{e}_{\vec{q}}\cdot\vec{e}_x \equiv 1$. It is possible to consider two qualitatively distinct classes of phonon spectra (a) isotropic with $\omega_q \approx cq$ and (b) anisotropic phonon spectrum (with cylindrical symmetry) $\omega_q = \sqrt{c_\parallel^2 q_\parallel^2 + c_\perp^2 q_\perp^2}$, where $q_\parallel$ denotes the component of the phonon quasimomentum directed along the chain, $q_\perp^2$ is the sum of the squares of the transverse components of phonon quasimomentum, while $c_\parallel$ and $c_\perp$ denote the longitudinal and transverse speed of sound, respectively.

We now proceed by applying adiabatic variational method[5,14] in combination with the scaling approach. This demands elimination of the phonon variables from the evolution equation for polaron wave function. It is possible if one uses a trial state assuming total separability of electron and phonon parts of the trial state $|\Psi\rangle = |\psi\rangle \otimes |\phi\rangle$. This is feasible in the adiabatic limit when vibrational subsystem may be treated essentially classically so that the phonon part of trial state may be chosen as multimode coherent state of phonon operators $|\phi\rangle = \Pi_q |\alpha_q\rangle$; $(a_q|\alpha_q\rangle = \alpha_q|\alpha_q\rangle)$, while $|\psi\rangle = \Sigma_n \psi_n B_n^\dagger |0\rangle$; $(\Sigma_n |\psi_n|^2 = 1)$ denotes the electron part of trial state. Functions ψ_n and α_q may be treated as dynamical variables satisfying the set of coupled evolution equations derived by means of time-dependent variational principle. Equation of motion for α_q may be solved easily; substituting the so-obtained result in the equation of motion for ψ_n and adopting the continuum approximation we obtain

$$\left[i\hbar\frac{\partial}{\partial t} + JR_0^2\frac{\partial^2}{\partial x^2} + G\int \frac{dx'}{R_0}\mathcal{K}(x-x')|\psi(x',t)|^2 \right]\psi(x,t) = 0. \quad (2)$$

Here $G = 4\chi^2 R_0^2/Mc_\parallel^2 \approx 2E_B$, where E_B denotes binding energy of the 1D small polaron which, together with the electron bandwidth ($2J$) and maximal phonon frequency (ω_B), determine the main features of electron-phonon systems. It is understood that $c_\parallel = c$ in the isotropic case. Kernel $\mathcal{K}(x)$ has the form

$$\mathcal{K}(x) = \frac{1}{N}\sum_{\vec{q}} \frac{c_\parallel^2 (\vec{q}\cdot\vec{e}_x)^4 e^{i\vec{q}\cdot\vec{e}_x x}}{q^2[\omega_q^2 - (\vec{q}\cdot\vec{v})^2]}. \quad (3)$$

Here v denotes the polaron velocity which appears due to the assumption $|\psi(x,t)|^2 = |\psi(x-vt)|^2$. This implies that the particular solution for the evolution equation for phonon coherent amplitudes has a simple form $\alpha_q(t) = e^{i\vec{q}\cdot\vec{v}t}\alpha_q(0)\{\alpha_q(0) = [F_q/\hbar(\omega_q - \vec{q}\cdot\vec{v})]\int dx e^{i\vec{q}\cdot\vec{e}_x x}|\psi(x)|^2\}$. In pure one-dimensional systems kernel approaches a δ-function form $\mathcal{K}(x) = \{1/[1 - (v/c)^2]\}R_0\delta(x)$, and the above nonlinear equation becomes the known nonlinear Schrödinger equation with local nonlinearity whose properties are well known. In the present case, however, it cannot be solved and is of no use in the examination of polaron properties. Fortunately, its explicit solution is not always necessary since the majority of the questions on the subject may be answered on the basis of simple qualitative analysis: the scaling method in analogy with the virial theorem. This method may be applied to a wide class of nonlinear partial equations which are of the Hamiltonian type[6–9,21] (i.e., they can be derived by demonstrating the stationarity of a certain nonlinear functional—the Hamiltonian). In general, Hamiltonian can be expressed as a sum of a few integrals, which are usually called kinetic energy, potential energy, total momentum, etc., in analogy with the classical mechanics. Scaling method enables one to find the relations between these integrals, which must be satisfied to ensure the stability of solutions.

In the adiabatic polaron theories this functional is simply the expectation value of model Hamiltonian (1) taken in the aforementioned trial state. After elimination of phonon variables it becomes

$$\mathcal{H} = JR_0^2\int \frac{dx}{R_0}\left|\frac{\partial\psi(x)}{\partial x}\right|^2 - \frac{G}{2}\int\frac{dx}{R_0}\frac{dx'}{R_0}\mathcal{K}(x-x') \times |\psi(x)|^2|\psi(x')|^2. \quad (4)$$

The first and second integrals in the above expressions are usually called kinetic and potential energy, respectively.

Let us now consider functions $\psi(x)$, which are stationary solutions of Eq. (2) and which satisfy the following scaling law: $\psi(x) \to \lambda^{1/2}\psi(\lambda x)$ when $x \to \lambda x$. Then, by virtue of the fact that stable solutions must satisfy conditions $\delta\mathcal{H} = 0$ and $\delta^2\mathcal{H} > 0$, it follows that $\partial\mathcal{H}/\partial\lambda|_{\lambda=1} = 0$ and $\partial^2\mathcal{H}/\partial\lambda^2|_{\lambda=1}$

>0. The first of these relations represents the condition that must be satisfied if ψ represents the stationary point of $\mathcal{H}$. Substitution of this relation into the second one gives a further condition that must be satisfied by the kinetic and potential energy in order for such stationary solutions to be stable. It is a necessary condition for the applicability of the above method that these integrals must converge when the integration is taken over all space. Localized, pulse-shaped functions characterized by some finite radius satisfy this criterion and are good candidates for variational treatment of the present problem when exact solutions of the relevant nonlinear partial differential equations are not known. Scaling arguments are still valid for such approximate treatments, where the scaling parameter may be related to the polaron radius.

III. DIMENSIONALITY OF PHONON SPECTRUM AND POLARON STABILITY

Let us now concentrate on the systems with 1D electronic spectrum embedded in realistic three-dimensional lattice. Two qualitatively distinct classes of phonon spectra, isotropic and anisotropic, will be analyzed.

(a) *Isotropic phonon spectrum*. In this case the sum over the phonon quasimomenta may be replaced by momentum space integration in accordance with the rule $(1/N)\Sigma_{\vec{q}} \rightarrow \int_0^{\pi} \sin\theta d\theta (3/2Q^3)\int_0^Q q^2 dq$. Here Q denotes phonon quasimomentum cutoff. In such a way, adiabatic functional, by virtue of auxiliary relation $\mathcal{K}(x-x')=(\partial^2/\partial x\partial x')\tilde{\mathcal{K}}(x-x')$ in which $\tilde{\mathcal{K}}(x)=(3/2Q^3)\int_{-1}^{1}[\mu^2 d\mu/(1-v^2/c^2\mu^2)]\int_0^Q e^{iq\mu x}dq$ ($\mu=\sin\theta$), may be written as

$$\mathcal{H}=JR_0^2\int\frac{dx}{R_0}\left|\frac{\partial\psi(x)}{\partial x}\right|^2-\frac{G}{2}\int\int\frac{dx}{R_0}\frac{dx'}{R_0}\times\tilde{\mathcal{K}}(x-x')\frac{\partial|\psi(x)|^2}{\partial x}\frac{\partial|\psi(x')|^2}{\partial x'}. \tag{5}$$

After the aforementioned norm-preserving scale change it becomes the function of scaling parameter $\mathcal{H}(\lambda)=\lambda^2E_k-\lambda^2E_p(\lambda)$. Here $E_p(\lambda)$ corresponds to the second term in the last equation in which $\tilde{\mathcal{K}}(x)$ is replaced by $\tilde{\mathcal{K}}(x/\lambda)$. In accordance with the continuum approximation, the upper limit of the "q" integration should be understood to be essentially infinite ($Q\rightarrow\infty$), in the sense that the basic assumption of the continuum approximation is that the phenomena of interest are insensitive to a structure on the scale of Q^{-1}. Consequently, after the variable change $q/\lambda=\tilde{q}$ and after extending the integration over "q" towards the infinity we have $\tilde{\mathcal{K}}\approx-(3\pi c^2/2Q^3v^2)\ln(1-v^2/c^2)\delta(x)$. This means that $\tilde{\mathcal{K}}(x/\lambda)\rightarrow\lambda\tilde{\mathcal{K}}(x)$ so that the adiabatic functional attains the following form:

$$\mathcal{H}(\lambda)=\lambda^2E_k-\lambda^3E_p. \tag{6}$$

This is precisely the same relation as the one found previously by Emin and Holstein[6] for truly 3D systems. Thus, even if the electronic subsystem is truly 1D, stable finite radius large polaron solution fails to exist if the host lattice is three dimensional and isotropic.

(b) *Anisotropic phonon spectrum*. At this stage it could be objected that the phonon isotropy is not to be expected in the systems where quasi-one-dimensional electronic propagation is found, since the most often quasi-one-dimensional nature of the electronic system is due to a structural anisotropy that should be reflected to some noticeable degree in the vibrational system. For that reason, we now consider the case in which the phonon system is three dimensional, but generally has less than spherical symmetry. Since the crucial issue hinges on the distinction between parallel and transverse axes, it is sufficient to consider the vibrational system to have cylindrical symmetry about the 1D electronic axes. Thus we use the following transcription, $(1/N)\Sigma_{\vec{q}}\rightarrow(1/2Q_{\parallel})\int_{-Q_{\parallel}}^{Q_{\parallel}}dq_{\parallel}(2/Q_{\perp}^2)\int_0^{Q_{\perp}}q_{\perp}dq_{\perp}$, where $Q_{\parallel}$ and $Q_{\perp}$ denote parallel and transverse phonon quasimomentum cutoff wave vectors, respectively. The relevant energy functional is substantially the same as Eq. (4); the only exception is in the form taken by the kernel $\mathcal{K}(x)$

$$\mathcal{K}(x)=\frac{1}{1-\dfrac{v^2+c_{\perp}^2}{c_{\parallel}^2}}\frac{1}{Q_{\parallel}Q_{\perp}^2}\int_{-Q_{\parallel}}^{Q_{\parallel}}q_{\parallel}^2dq_{\parallel}e^{iq_{\parallel}x}\times\left[\int_0^{Q_{\perp}}\frac{q_{\perp}dq_{\perp}}{q_{\parallel}^2+q_{\perp}^2}-\int_0^{\tilde{Q}_{\perp}}\frac{\tilde{q}_{\perp}d\tilde{q}_{\perp}}{q_{\parallel}^2+\tilde{q}_{\perp}^2}\right],\quad \tilde{q}_{\perp}=\frac{\dfrac{c_{\perp}}{c_{\parallel}}q_{\perp}}{\sqrt{1-\dfrac{v^2}{c_{\parallel}^2}}}. \tag{7}$$

In considering the effect of phonon anisotropy, we must pay some attention to the different values which may be associated with the above introduced cut-off wave vectors. We would like to have a result in which 3D isotropic case and pure 1D case might be realized as limits. The natural way to recover 1D result is to allow the interchain distance (b) to become infinite relative to the lattice constant within the 1D chains; in this limit $Q_{\perp}\sim\pi/b$ would vanish with respect to $Q_{\parallel}\sim\pi/R_0$. This limit, as can be seen from Eq. (7), can be meaningful in a final result only if the integration limit $Q_{\perp}$ should be kept finite and only $Q_{\parallel}$ is considered to be essentially infinite. In such a way the integrals in the last expression may be evaluated easily and we have

$$\mathcal{K}(x)\approx\frac{1}{1-\dfrac{v^2}{c_{\parallel}^2}}R_0\delta(x)-\frac{1}{1-\dfrac{v^2+c_{\perp}^2}{c_{\parallel}^2}}\frac{R_0}{Q_{\perp}^2}\frac{\partial^2}{\partial|x|^2}\frac{1}{|x|}\times(e^{-\tilde{Q}_{\perp}|x|}-e^{-Q_{\perp}|x|}). \tag{8}$$

In the above expressions we use symbol "$\approx$" as a reminder that in obtaining these results we have considered the integration limit $Q_{\parallel}$ to be essentially infinite, which introduces no significant errors since we are concerned with the regime in which $Q_{\parallel}\gg Q_{\perp}$.

The δ-function component produces potential energy term having one-dimensional scaling properties $[E_P'\sim(G/2)\int dz|\psi(z)|^4]$; the non-$\delta$ component produces a nonlocal potential energy term

$$E_P^{\mathrm{nl}} = \frac{G}{2}\left(\frac{\xi}{\pi}\right)^2 \int\int dz dz' \frac{\partial^2 \tilde{\mathcal{K}}(z-z')}{\partial(|z-z'|)^2} |\psi(z)|^2 |\psi(z')|^2. \quad (9)$$

Here we have introduced, for convenience, dimensionless variable $z=x/R_0$ while

$$\tilde{\mathcal{K}}(z) = \frac{1}{1-\dfrac{v^2+c_\perp^2}{c_\parallel^2}} \frac{1}{|z|} \left(e^{-(c_\perp/c_\parallel)(|z|/\xi)} - e^{-(|z|/\xi)}\right). \quad (10)$$

It accounts for the nonlocality of the electron-phonon interaction whose range is determined by the magnitude of correlating length $\xi=\pi Q_\parallel/Q_\perp (\pi<\xi<\infty)$. Performing the norm preserving scaling transformation we obtain

$$\mathcal{H}(\lambda) = \lambda^2 E_K - \lambda E_P^l + \lambda^3 E_P^{\mathrm{nl}}(\lambda). \quad (11)$$

Here $E_P^{\mathrm{nl}}(\lambda)$ denotes nonlocal potential energy Eq. (9) with scaled correlation length $\xi(\lambda)=\lambda\xi$.

Consequences of this term to the large polaron properties are determined by the behavior of $\tilde{\mathcal{K}}(z-z')$ as a function of distance $|z-z'|$, which substantially depends on interaction range (ξ). This function is not as singular as it may appear. It has a fixed, finite area whose size is independent of the value of ξ which, however, determines its height and width. For large ξ, i.e., $\xi \gg |z-z'|$, it is slowly varying and extended over a large area. In the opposite case it is highly peaked and very narrow rapidly decaying at large distances. Scale change is equivalent to a contraction of correlation length $(0 \leqslant \lambda \leqslant 1)$, which results in shrinking of $\tilde{\mathcal{K}}(z;\lambda)$ and in the increase of its height. This is especially expressed in the short correlation length limit when even small scale change significantly affects $\tilde{\mathcal{K}}(z;\lambda)$, which resembles a δ function. In this limit the last term in Eq. (11) scales as, approximately, λ^3 and $\mathcal{H}(\lambda)$ has always stable minimum which, in respect to the pure 1D case, moves toward smaller values of λ and the enlarging of polaron size arises. We are, however, concerned with long-range limit $(\xi \to \infty)$ and therefore the above conclusion should be taken with some reserves. In this limit $\tilde{\mathcal{K}}(z)$ is broad, practically flat, and independent on λ. In this case we may perform an approximate calculation expanding the exponentials in Eq. (10) in powers of "small parameter" $\alpha|z-z'|$; $(\alpha=(c_\perp/c_\parallel)/\xi; 1/\xi)$. In such a way in a static limit $(v=0)$ we found

$$\mathcal{H}(\lambda) \approx \lambda^2 E_K - \lambda E_P^l + \frac{\pi G}{6}\frac{Q_\perp}{Q_\parallel} \frac{1+\dfrac{c_\perp}{c_\parallel}+\left(\dfrac{c_\perp}{c_\parallel}\right)^2}{1+\dfrac{c_\perp}{c_\parallel}}. \quad (12)$$

Thus, the correction due to the transverse dimensions amounts to only a structureless shift increase in the ground state energy ($E_{\mathrm{g.s.}}$), which does not scale but may violate polaron stability if this shift exceeds the ground state energy of pure 1D systems.

Requiring $E_{\mathrm{g.s.}}<0$ we found the stability condition for large polarons in these media

$$\frac{E_B}{2J} > 2\pi \frac{Q_\perp}{Q_\parallel} \frac{1+\dfrac{c_\perp}{c_\parallel}+\left(\dfrac{c_\perp}{c_\parallel}\right)^2}{1+\dfrac{c_\perp}{c_\parallel}}. \quad (13)$$

In approaching this result we have evaluated the integrals in Eq. (12) taking the polaron wave function in the form $\psi(z) = \sqrt{(\mu/2)}\,\mathrm{sech}\,\mu z$, which minimizes functional (12) for $\lambda=1$ and $\mu=E_B/2J$. The ratio on the left-hand side of Eq. (13) is proportional to the reciprocal large polaron radius measured in units of lattice constant along the chain and, consequently, the last relation implies that the polaron size cannot exceed some critical value determined by the ratio $Q_\parallel/Q_\perp \sim b/R_0$. Introducing dimensionless polaron radius $l=l_{\mathrm{pol}}/R_0$, where $l_{\mathrm{pol}} \sim \pi R_0/\mu$ (Ref. 14) denotes radius of 1D soliton (polaron), we estimate this critical size as $l_c \sim b/2R_0$ and now large polaron stability condition reads $l<b/(2R_0)$. On the other hand, applicability of the continuum approximation requires polaron spacing to be much larger than the intrachain lattice constant which, together with the above established criterion, imposes $1<l<b/(2R_0)$. Having in mind the connection of the soliton radius and physical parameters of system, from this criterion we conclude that the large polaron existence in these systems imposes the following restriction on the values of basic system parameters: $1<2J/E_B<b/2R_0$.

We finally emphasize that a very similar result has been obtained by Schüttler and Holstein,[23] who found that, due to the interchain coupling, polaron would become unstable if its size exceeds critical limit $L_c=(c_\parallel/c_\perp)b$. In contrast to the present model they have considered both transverse electronic transfer and elastic interchain coupling.

IV. CONCLUDING REMARKS

In summary we underline that our aim in this paper has been to address the conflict between the theoretical ideal of one-dimensional behavior and the physical reality of three-dimensional solids. We have dismissed from consideration the inevitable presence of small transverse hopping integrals[8,9,23] in order to focus on the manner in which the dimensionality of the host solid affects the electron (exciton)-phonon interaction; we found that even when the direct electron-phonon interaction is confined to a single chain, indirect effects due to the disturbances transverse to the chain have a considerable impact on the adiabatic state of the excitation within the chain.

Our major result is that the transition from one-dimensional to three-dimensional behavior occurs as a function of the ratio $Q_\parallel/Q_\perp$. In the isotropic solids, this ratio is unity, implying that there is no meaningful length scale on which one-dimensional polaron can exist. In anisotropic solids, this ratio may be greater than unity, allowing one-dimensional correlations to persist over a few lattice sites, but there is no real solid in which this ratio can be excessively large. We must conclude from this behavior that the large polaron radius, even in solids argued to be quasi-one-dimensional in their electronic structure, must be limited and

can extend over a few lattice sites only. In that respect our results are similar to those of Emin,[9] and Schüttler and Holstein,[23] who found that the interchain coupling, in the systems with short-ranged interaction of an electron with dispersionless optical[9] and longitudinal acoustic modes,[23] enforces the analogous restrictions on the upper bound for large polaron spatial extent. However, having in mind that the present analysis concerns ideal 1D electronic systems, our criterion imposes even stronger restrictions on large polaron existence since it emphasizes that, even when transverse electron transfer may be neglected, dimensionality of phonon spectra may seriously affect large polaron stability.

ACKNOWLEDGMENTS

We would like to acknowledge useful conversations with Dr. Katja Lindenberg, Dr. Darko Kapor, and Dr. Slobodan Zeković. This work was partially supported by the Serbian Ministry of Science and Technology under Contract No. 1961.

*Electronic address: zivic@rt270.vin.bg.ac.yu

[1] *Lattice Effects in High-T_c Superconductors*, edited by Y. Bar-Yam, T. Egami, J. Mustre de Leon, and A. R. Bishop (World Scientific, Singapore, 1992); A. S. Alexandrov and N. F. Mott, *Polarons and Bipolarons* (World Scientific, Singapore, 1995).

[2] G. M. Zhao, K. Conder, H. Keller, and K. A. Muller, Nature (London) **381**, 676 (1996); H. Röder, J. Zang, and A. R. Bishop, Phys. Rev. Lett. **76**, 1356 (1996); A. Lanzara, N. L. Saini, M. Brunelli, F. Natali, A. Bianconi, P. G. Radaelli, and S. W. Cheong, *ibid.* **81**, 878 (1998).

[3] A. J. Heeger, S. Kivelson, and J. R. Schriefer, Rev. Mod. Phys. **60**, 781 (1988); S. A. Brazovskii, JETP Lett. **28**, 606 (1978); W. P. Su, J. R. Schrieffer, and A. J. Heeger, Phys. Rev. Lett. **42**, 1698 (1979).

[4] E. M. Conwell and S. V. Rakhmanova, Proc. Natl. Acad. Sci. U.S.A. **97**, 4556 (2000); S. V. Rakhmanova and E. M. Conwell, J. Phys. Chem. B **105**, 2056 (2001).

[5] S. I. Pekar, *Issledovanaia po Elektronnoi Teorii Kristalov* (Gosteh, izd., Moskva, 1951).

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

[7] E. I. Rashba, Izv. Akad. Nauk SSSR, Ser. Fiz. **40**, 1793 (1976); E. I. Rashba and M. Struge, *Excitons* (North-Holland, Amsterdam, 1982).

[8] B. Pertzch and U. Rössler, Solid State Commun. **37**, 931 (1981).

[9] D. Emin, Phys. Rev. B **33**, 3973 (1986).

[10] H. Sumi and A. Sumi, J. Phys. Soc. Jpn. **63**, 637 (1994).

[11] A. H. Romero, D. W. Brown, and K. Lindenberg, Phys. Rev. B **60**, 14 080 (1999); **60**, 4618 (1999); A. Shelkan, V. Hizhnyakov, and E. Sigmund, J. Supercond. **104**, 21 (2000).

[12] M. Zoli, Phys. Rev. B **61**, 14 523 (2000), J. Phys.: Condens. Matter **13**, 2437 (2001).

[13] Li-Chung Ku, S. A. Trugman, and J. Bonča, Phys. Rev. B **65**, 174306 (2002).

[14] A. S. Davydov, Usp. Fiz. Nauk **138**, 603 (1982) [Sov. Phys. Usp. **25**, 898 (1982)].

[15] D. Jerome, J. Phys. (Paris), Colloq. **44**, 775 (1983); D. Jerome and H. J. Schulz, Adv. Phys. **31**, 229 (1982).

[16] L. Rothberg, T. M. Jedju, P. D. Townsend, S. Etemad, and G. L. Baker, Phys. Rev. Lett. **65**, 100 (1990).

[17] M. Sinclair, D. Moses, and A. J. Heeger, Solid State Commun. **59**, 343 (1981).

[18] Y. Furukawa, A. Sakamoto, H. Ohta, and M. Tasumi, Synth. Met. **49**, 335 (1992).

[19] H. A. Mizes and E. M. Conwell, Phys. Rev. Lett. **70**, 1505 (1993).

[20] A. A. Gogolin, Solid State Commun. **50**, 265 (1984).

[21] E. A. Kuznetsov, A. M. Rubenchik, and V. E. Zaharov, Phys. Rep. **142**, 103 (1986).

[22] D. M. Alexander and J. A. Krumhansl, Phys. Rev. B **33**, 7172 (1986).

[23] H. Schütler and T. Holstein, Ann. Phys. (N.Y.) **166**, 93 (1986).