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Polaron mobility obtained by a variational approach for lattice Fröhlich models

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ABSTRACT

Charge carrier mobility for a class of lattice models with long-range electron–phonon interaction was investigated. The approach for mobility calculation is based on a suitably chosen unitary transformation of the model Hamiltonian which transforms it into the form where the remaining interaction part can be treated as a perturbation. Relevant spectral functions were then obtained using Matsubara Green's functions technique and charge carrier mobility was evaluated using Kubo's linear response formula. Numerical results were presented for a wide range of electron–phonon interaction strengths and temperatures in the case of one-dimensional version of the model. The results indicate that the mobility decreases with increasing temperature for all electron–phonon interaction strengths in the investigated range, while longer interaction range leads to more mobile carriers.

1. Introduction

There is a strong interest to understand the effects of electron–phonon interaction on electrical transport properties of semiconductors since it is the interaction mechanism that is present in every material being in most cases the dominant mechanism that limits the charge carrier mobility. In the case of semiconductors with wide bands and weak electron–phonon interaction (such as for example conventional inorganic semiconductors GaAs or Si), charge carrier mobility can be described using Bloch–Boltzmann theory [1–3] and it can be evaluated from electron–phonon scattering time and the effective mass of the carrier. Significant research efforts are currently devoted towards developing ab-initio methods for calculation of mobility in this regime [4–10] which is a non-trivial task due to the

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necessity of taking a large number of points in the Brillouin zone to obtain accurate values of scattering times [11,12] and due to the difficulties of including long-range interaction with polar optical phonons within such approach [13,14]. In the opposite limit of narrow bands and strong electron–phonon interaction (which can be valid in some organic semiconductors based on small molecules) charge carrier transport is typically modeled using hopping theories [15], with hopping rates between the molecules evaluated using Marcus formula [16,17] or its generalizations [18–20], which consider only local electron–phonon interaction.

It is of significant importance to develop methods that can be used to evaluate charge carrier mobility beyond these two limiting regimes [21–24]. Due to difficulties in treating electron–phonon interaction of intermediate strength, the efforts to develop such methods are more scarce. These methods are usually based on a unitary transformation of the model Hamiltonian [25,26] and in practice these are applied to Hamiltonians with local electron–phonon interaction (Holstein model) [26,25] or short-range non-local interaction [27–29] (Peierls model). More recently, Quantum Monte Carlo techniques were also applied to evaluate the mobility in the Holstein and the Peierls model [30,31].

The main aim of this work is to develop a method for evaluating the mobility in systems with long-range electron–phonon interaction for a wide range of interaction strengths and temperatures. We consider a lattice model with long-range electron–phonon interaction of Fröhlich type and evaluate the mobility using an approach that combines unitary transformation of the Hamiltonian, Matsubara Green's function technique for evaluation of relevant spectral functions and Kubo's formula for calculation of mobility. The manuscript is organized as follows. In Section 2 we introduce the model Hamiltonian that is the subject of this work. In Section 3 we present the unitary transformation that is used to transform the Hamiltonian to the form where the remaining interaction can be treated using perturbative techniques, we derive the equations for optimal parameters of the unitary transformation and present numerical results for bandwidth renormalization. Equations for self-energies obtained using Matsubara Green's function technique are presented in Section 4 along with the numerical results for polaronic spectral functions. In Section 5 we present the derivation of mobility based on Kubo's linear response theory and numerical results for a wide range of temperatures and electron–phonon coupling strengths. Concluding remarks are given in Section 6.

2. Model Hamiltonian

We consider the following Hamiltonian that describes a periodic system of electrons and phonons that interact via long-range interaction:

$$H = -\sum_{\mathbf{m},\mathbf{n}} t_{\mathbf{m}-\mathbf{n}} c_{\mathbf{m}}^{\dagger} c_{\mathbf{n}} + \sum_{\mathbf{n}} \hbar\omega_0 b_{\mathbf{n}}^{\dagger} b_{\mathbf{n}} - \\ - \sum_{\mathbf{m},\mathbf{n}} \hbar\omega_0 f_{\mathbf{m}-\mathbf{n}} c_{\mathbf{n}}^{\dagger} c_{\mathbf{n}} (b_{\mathbf{m}}^{\dagger} + b_{\mathbf{m}}). \tag{1}$$

In Eq. (1) the vectors $\mathbf{m}$ and $\mathbf{n}$ label the sites of an infinite lattice, $c_{\mathbf{m}}$, $c_{\mathbf{m}}^{\dagger}$, $b_{\mathbf{m}}$, and $b_{\mathbf{m}}^{\dagger}$ are respectively annihilation and creation operators for electrons and phonons, $t_{\mathbf{m}-\mathbf{n}}$ is the electronic transfer integral that quantifies the electronic coupling between sites $\mathbf{m}$ and $\mathbf{n}$, $\hbar\omega_0$ is the energy of a phonon, while $f_{\mathbf{m}-\mathbf{n}}$ are dimensionless electron–phonon coupling parameters.

The Hamiltonian H considers a single electronic state per period of the system and a single dispersionless phonon band. While accurate modeling of real materials would certainly require a more elaborate Hamiltonian, we believe that it is essential first to address the properties of this relatively simple Hamiltonian. In the case when coupling parameters $f_{\mathbf{m}-\mathbf{n}}$ are zero except for $\mathbf{m} = \mathbf{n}$ the Hamiltonian reduces to a widely studied Holstein Hamiltonian [32]. In this work, we address a rather different scenario when $f_{\mathbf{m}-\mathbf{n}}$ is long-ranged.

Long-ranged electron–phonon interaction occurs as a consequence of interaction of electrons with a polarization field created from optical phonons in a polar material. To be more specific, an optical phonon at site $\mathbf{m}$ creates a dipole moment $\mathbf{p}_{\mathbf{m}} = p_{\mathbf{m}}\mathbf{e}_{\mathbf{m}}$, where $p_{\mathbf{m}}$ is its intensity and $\mathbf{e}_{\mathbf{m}}$ the unit vector in the direction of the dipole. Classical electrical potential at site $\mathbf{n}$ created by this dipole