Femtosecond Evolution of Spatially Inhomogeneous Carrier Excitations Part I: Kinetic Approach

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Abstract. The ultrafast evolution of optically excited carriers which propagate in a quantum wire and interact with three dimensional phonons is investigated. The equation, relevant to this physical problem, is derived by a first principle approach. The electron-phonon interaction is described on a quantum-kinetic level by the Levinson equation, but the evolution problem becomes inhomogeneous due to the spatial dependence of the initial condition. The initial carrier distribution is assumed Gaussian both in energy and space coordinates, an electric field can be applied along the wire. A stochastic method, described in Part II of the work, is used for solving the equation. The obtained simulation results characterize the space and energy dependence of the evolution in the zero field case. Quantum effects introduced by the early time electron-phonon interaction are analyzed.

1 The Coupled Electron-Phonon System

We consider a system of electrons which interact with the lattice vibrations. The electric forces which accelerate the electrons are due to the structure potential and the applied bias, Coulomb interaction between the electrons is neglected. The description of the system is provided by both the electron and the phonon degrees of freedom. We derive the Wigner equation for the coupled electron-phonon system. The corresponding Hamiltonian is given by the free electron part H_0, the structure potential $V(\mathbf{r})$, the free-phonon Hamiltonian H_p, and the electron-phonon interaction H_{e-p}:

$$H = H_0 + V + H_p + H_{e-p} = \\ -\frac{\hbar^2}{2m}\nabla_{\mathbf{r}} + V(\mathbf{r}) + \sum_{\mathbf{q}} b^{\dagger}_{\mathbf{q}} b_{\mathbf{q}} \hbar\omega_{\mathbf{q}} + i\hbar \sum_{\mathbf{q}} \mathcal{F}(\mathbf{q})(b_{\mathbf{q}} e^{i\mathbf{qr}} - b^{\dagger}_{\mathbf{q}} e^{-i\mathbf{qr}}) \quad (1)$$

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Here $b^{\dagger}_{\mathbf{q}}$ and $b_{\mathbf{q}}$ are the creation and annihilation operators for the phonon mode $\mathbf{q}$, $\omega_{\mathbf{q}}$ is the energy of that mode, and $\mathcal{F}(\mathbf{q})$ is the electron-phonon coupling element, which depends on the type of phonon scattering analyzed. The state of the phonon subsystem is presented by the set $\{n_{\mathbf{q}}\}$ where $n_{\mathbf{q}}$ is the occupation number of the phonons in mode $\mathbf{q}$. The representation of the basis set is provided by the vectors $|\{n_{\mathbf{q}}\}, \mathbf{r}\rangle = |\{n_{\mathbf{q}}\}\rangle|\mathbf{r}\rangle$.

The considered structure is a quantum wire, formed by potential barriers which confine the electron system in the plane normal to the wire. In this plane, at low temperatures, the system occupies the ground state Ψ. A homogeneous electric field E can be applied along the direction of the wire z. It holds:

$$H_0 + V(\mathbf{r}) = H_{\perp} + H_z = H_{0\perp} + V_{\perp} + H_{0z} + V(z); \qquad H_{\perp}\Psi = E_{\perp}\Psi,$$

$V(z) = -eEz$ and $|\mathbf{r}\rangle = |\mathbf{r}_{\perp}\rangle|z\rangle$. The generalized electron-phonon Wigner function is defined by the Fourier transform of the density operator $\hat{\rho}$:

$$f_w(z, p_z, \{n_{\mathbf{q}}\}, \{n'_{\mathbf{q}}\}, t) = \int \frac{d\mathbf{r}'}{2\pi\hbar} e^{-ip_z z'/\hbar} \langle z + \frac{z'}{2}, \{n_{\mathbf{q}}\}|\langle \mathbf{r}'_{\perp}|\hat{\rho}_t|\mathbf{r}'_{\perp}\rangle|\{n'_{\mathbf{q}}\}, z - \frac{z'}{2}\rangle$$

The evolution problem is separated with respect to the normal and z coordinates as follows:

$$\hat{\rho} = |\Psi\rangle\langle\Psi|\hat{\rho}_{tz}; \qquad \langle \mathbf{r}, \{n_{\mathbf{q}}\}|\hat{\rho}_t|\{n'_{\mathbf{q}}\}, \mathbf{r}'\rangle = \Psi^*(\mathbf{r}'_{\perp})\Psi(\mathbf{r}_{\perp})\rho(z, z', \{n_{\mathbf{q}}\}, \{n'_{\mathbf{q}}\}, t)$$

Assuming that Ψ is normalized to unity it is obtained:

$$f_w(z, p_z, \{n_{\mathbf{q}}\}, \{n'_{\mathbf{q}}\}, t) = \int \frac{dz'}{2\pi\hbar} e^{-ip_z z'/\hbar} \rho(z + \frac{z'}{2}, z - \frac{z'}{2}, \{n_{\mathbf{q}}\}, \{n'_{\mathbf{q}}\}, t)$$

The equation of motion of f_w is obtained from the von-Neumann equation for the density matrix:

$$\frac{\partial f_w(z, p_z, \{n_{\mathbf{q}}\}, \{n'_{\mathbf{q}}\}, t)}{\partial t} =$$
$$\frac{1}{i2\pi\hbar^2} \int dz' \int d\mathbf{r}_{\perp} e^{-ip_z z'/\hbar} \langle z + \frac{z'}{2}, \{n_{\mathbf{q}}\}|\langle \mathbf{r}_{\perp}|\,[H, \hat{\rho}_t]_-\,|\mathbf{r}_{\perp}\rangle|\{n'_{\mathbf{q}}\}, z - \frac{z'}{2}\rangle$$

The right hand side, evaluated for each term of the Hamiltonian (1) gives rise to the generalized Wigner equation for the confined electron system. The equation couples an element $f_w(..., \{n\}, \{m\}, t)$ to four neighborhood elements for any phonon mode $\mathbf{q}$. For any such mode $n_{\mathbf{q}}$ can be any integer between 0 and infinity and a sum over $\mathbf{q}'$ couples all modes. Of interest is the reduced or electron Wigner function defined as the trace over the phonon coordinates:

$$f_w(z, p_z, t) = \sum_{\{n_{\mathbf{q}}\}} f_w(z, p_z, \{n_{\mathbf{q}}\}, \{n_{\mathbf{q}}\}, t)$$

A closed equation for the reduced Wigner function is obtained by a set of assumptions and approximations: A diagonal element is linked to elements, called first

off-diagonal elements, which are diagonal in all modes but the current mode $\mathbf{q}'$ of the summation. In this mode the four neighbors of $n_{\mathbf{q}'}, n_{\mathbf{q}'}$ namely $n_{\mathbf{q}'}\pm 1, n_{\mathbf{q}'}$ and $n_{\mathbf{q}'}, n_{\mathbf{q}'}\pm 1$ are concerned. For convenience we denote the phonon state, obtained from $\{n_{\mathbf{q}}\}$ by increasing or decreasing the phonons in mode $\mathbf{q}'$ by unity as $\{n_{\mathbf{q}}\}^{\pm}_{\mathbf{q}'}$. The first off-diagonal elements are linked to elements which in general are placed further away from the diagonal ones by increasing or decreasing the phonon number in a second mode, $\mathbf{q}''$, by unity. These are the second off-diagonal elements. The only exception is provided by two contributions which recover diagonal elements. They are obtained from $(\{\{n_{\mathbf{q}}\}^{+}_{\mathbf{q}'}\}^{-}_{\mathbf{q}''}, \{n_{\mathbf{q}}\})$ and $(\{n_{\mathbf{q}}\}^{+}_{\mathbf{q}'}, \{n_{\mathbf{q}}\}^{+}_{\mathbf{q}''})$ in the case when the two phonon modes coincide: $\mathbf{q}' = \mathbf{q}''$. The first approximation is to keep only such terms in the equations for the first off-diagonal terms. Then the equations for the diagonal and the four first off-diagonal terms form a closed system. Furthermore, the four first off-diagonal equations can be solved and substituted in the diagonal one. The obtained equation contains only diagonal terms such as $f_w(z, p_z, \{n_{\mathbf{q}}\}, \{n_{\mathbf{q}}\}, t)$ and $f_w(z, p_z, \{n_{\mathbf{q}}\}^{\pm}_{\mathbf{q}'}, \{n_{\mathbf{q}}\}^{\pm}_{\mathbf{q}'}, t)$. The next major assumption is that the phonon system remains in equilibrium during the evolution. This allows to take the trace on the phonon coordinates and to obtain an equation for the reduced Wigner function $f_w(z, p_z, t)$. After few steps of transformations, which include the settings

$$\sum_{\mathbf{q}'} = \frac{V}{(2\pi)^3}\int d\mathbf{q}'; \qquad k_z = p_z/\hbar; \qquad k'_z = k_z - q_z$$

and a conversion to an integral form, the equation reads:

$$\begin{aligned} f(z,k_z,t) = f(z(0),k_z(0),0) + \int_0^t \mathrm{d}t' \int_0^{t'} \mathrm{d}t'' \int d\mathbf{q}'_\perp \int dk'_z \times \qquad (2)\\ \Big[S(k'_z,k_z,t',t'',\mathbf{q}'_\perp) f(z(t'') + \frac{\hbar q'_z}{2m}(t'-t''), k'_z(t''), t'') - \\ S(k_z,k'_z,t',t'',\mathbf{q}'_\perp) f(z(t'') + \frac{\hbar q'_z}{2m}(t'-t''), k_z(t''), t'')\Big] \end{aligned}$$

$$\begin{aligned} S(k'_z,k_z,t',t'',\mathbf{q}'_\perp) = \frac{2V}{(2\pi)^3}|G(\mathbf{q}'_\perp)\mathcal{F}(\mathbf{q}'_\perp, k_z - k'_z)|^2 \times \\ \Bigg[(n(\mathbf{q}')+1) cos\left(\int_{t''}^{t'} \mathrm{d}\tau \frac{1}{\hbar}\Big(\epsilon(k_z(\tau)) - \epsilon(k'_z(\tau)) + \hbar\omega_{\mathbf{q}'}\Big)\right) \\ + \; n(\mathbf{q}') cos\left(\int_{t''}^{t'} \mathrm{d}\tau \frac{1}{\hbar}\Big(\epsilon(k_z(\tau)) - \epsilon(k'_z(\tau)) - \hbar\omega_{\mathbf{q}'}\Big)\right)\Bigg] \end{aligned}$$

Equation 2 generalizes the Levinson equation [1] for the case of a space dependent initial condition. The Newton trajectories, initialized at z, k_z, t, are governed by the electric force F:

$$z(t'') = z - \frac{1}{m}\int_{t''}^{t} p_z(\tau)d\tau; \qquad k_z(t'') = k_z - F(t-t''); \qquad F = \frac{eE}{\hbar} \qquad (3)$$

The shape of the wire affects the electron-phonon coupling through the factor G:

$$G(\mathbf{q}'_\perp) = \int d\mathbf{r}_\perp e^{i\mathbf{q}'_\perp \mathbf{r}_\perp} |\Psi(\mathbf{r}_\perp)|^2$$

2 Phase Space Transform

The equation reveals a very inconvenient from a numerical point of view property, namely that a solution for a phase space point (z, k_z) at t is linked with the solutions on the trajectories (3). Thus the simulation domain grows with the force F and the evolution time t in both position and wave vector subspaces. The following transform $(k_{z1} = k_z, k'_z)$ is suggested to cope with this problem:

$$k^t_{z1} = k_{z1} - Ft; \quad k_{z1}(\tau) = k^t_{z1} + F\tau; \quad f(z, k_z, t) = f(z, k^t_z + Ft, t) \stackrel{\text{def}}{=} f^t(z, k^t_z, t)$$

The following equalities can be easily shown:

$$f(z, k_z(t''), t'') = f(z, k^t_z + Ft'', t'') = f^t(z, k^t_z, t'')$$

$$z(t'') = z - \left(\frac{\hbar k^t_z}{m} + \frac{\hbar F}{2m}(t + t'') \right)(t - t'')$$

and $q'_z = k_z - k'_z = k^t_z - k'^t_z$.

$$\epsilon(k'_z(\tau)) - \epsilon(k_z(\tau)) = \epsilon(k'^t_z) - \epsilon(k^t_z) - \frac{2\hbar^2}{2m} F.q'_z \tau$$

All terms in the equation are now expressed as functions of k^t_z, k'^t_z. By omitting the superscripts of the arguments it is obtained:

$$f(z, k_z, t) = f(z - \frac{\hbar k_z}{m}t + \frac{\hbar F}{2m}t^2, k_z, 0) + \int_0^t \mathrm{d}t'' \int_{t''}^t \mathrm{d}t' \int d\mathbf{q}'_\perp \int dk'_z \times \qquad (4)$$
$$\Big[S(k'_z, k_z, t', t'', \mathbf{q}'_\perp) f \left(z - \frac{\hbar k_z}{m}(t - t'') + \frac{\hbar F}{2m}(t^2 - t''^2) + \frac{\hbar q'_z}{2m}(t' - t''), k'_z, t'' \right)$$
$$- S(k_z, k'_z, t', t'' \mathbf{q}'_\perp) f \left(z - \frac{\hbar k_z}{m}(t - t'') + \frac{\hbar F}{2m}(t^2 - t''^2) - \frac{\hbar q'_z}{2m}(t' - t''), k_z, t'' \right) \Big]$$

$$S(k'_z, k_z, t', t'', \mathbf{q}'_\perp) = \frac{2V}{(2\pi)^3} |G(\mathbf{q}'_\perp) \mathcal{F}(\mathbf{q}'_\perp, k_z - k'_z)|^2 \times$$
$$\Big[(n(\mathbf{q}') + 1) cos \left(\frac{\epsilon(k_z) - \epsilon(k'_z) + \hbar\omega_{\mathbf{q}'}}{\hbar}(t' - t'') + \frac{\hbar}{2m} F.q'_z (t'^2 - t''^2) \right)$$
$$+ n(\mathbf{q}') cos \left(\frac{\epsilon(k_z) - \epsilon(k'_z) - \hbar\omega_{\mathbf{q}'}}{\hbar}(t' - t'') + \frac{\hbar}{2m} F.q'_z (t'^2 - t''^2) \right) \Big]$$

3 Simulation Results and Discussions

We first consider the numerical properties of (4). The integration is in the wave vector space, while the real space variable enters as a parameter modified by the two time integrals. The advantage of (4) as compared to (2) is that the wave vector variable is decoupled from the time variable and thus the integration domain can be maintained independent of the force and the evolution time. Despite that the numerical challenges posed by (4) are heavy already in the homogeneous problem [2]. In the latter case one of the time integrals can be assigned to S. Furthermore another integration in the wave vector space can be spared due to symmetry considerations. In the inhomogeneous problem it is no more possible to assign the time integral to S due to the t' dependence of f in the right hand side of (4). The physical origin of this dependence is associated with the finite duration of the electron-phonon interaction: the real space trajectory is modified by the half of the phonon wave vector q'_z times the duration $t' - t''$ of the interaction. Thus in the general case each iteration step increases the dimensionality by five more integrals and thus the computational burden.

The equation accounts for interesting quantum effects demonstrated by the presented simulation results. Considered is a $GaAs$ material with a single polar optical phonon having a constant energy $\hbar\omega$. The electric field is zero. The initial condition is a product of two Gaussian distributions of the energy and space. The k_z^2 distribution corresponds to a generating laser pulse with an excess energy of about $150meV$. The z distribution is centered around zero. A quantum wire with a rectangular cross section is assumed. At very low temperature the physical system has a transparent semiclassical behavior. We recall the major results of the homogeneous case [2]. Semiclassical electrons can only emit phonons and loose energy equal to a multiple of the phonon energy $\hbar\omega$. They evolve according to an energy distribution, patterned by replicas of the initial condition shifted towards low energies. Such electrons cannot appear in the region above the initial distribution. The quantum solutions demonstrate two effects of deviation from the semiclassical behavior. The replicas are broadened and the broadening reduces with the time. A finite density of electrons appears in the semi-classically forbidden region above the initial condition. These effects are due to the lack of the energy conserving delta function, which is build up by the cosine function in S for long evolution times.

In the inhomogeneous case the wave vector (and respectively the energy) and the density distributions are given by the integrals

$$f(k_z, t) = \int \frac{dz}{2\pi} f_w(z, k_z, t); \qquad n(z, t) = \int \frac{dk_z}{2\pi} f_w(z, k_z, t)$$

Figure 1 shows the redistribution of the initial electrons after 50 femtoseconds evolution time as a function of the proportional to the energy quantity k_z^2. A window of values for k_z^2 and f is chosen, where the broadening of first replica and the finite density of electrons with energies above the initial condition is well visible. Figure 2 shows the distribution in the whole simulation domain after 150 femtoseconds evolution. The first replica becomes sharper, but still broadened

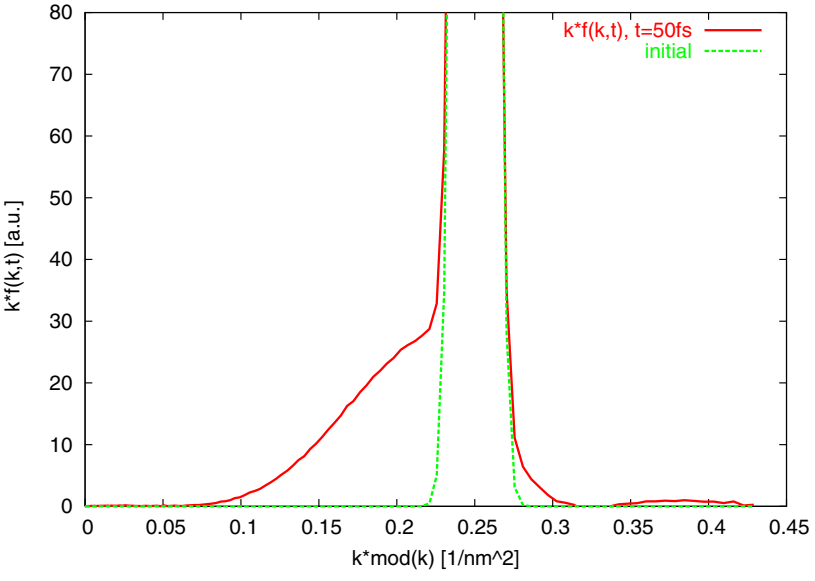

Fig. 1. Initial condition and energy distribution at $50fs$ evolution time

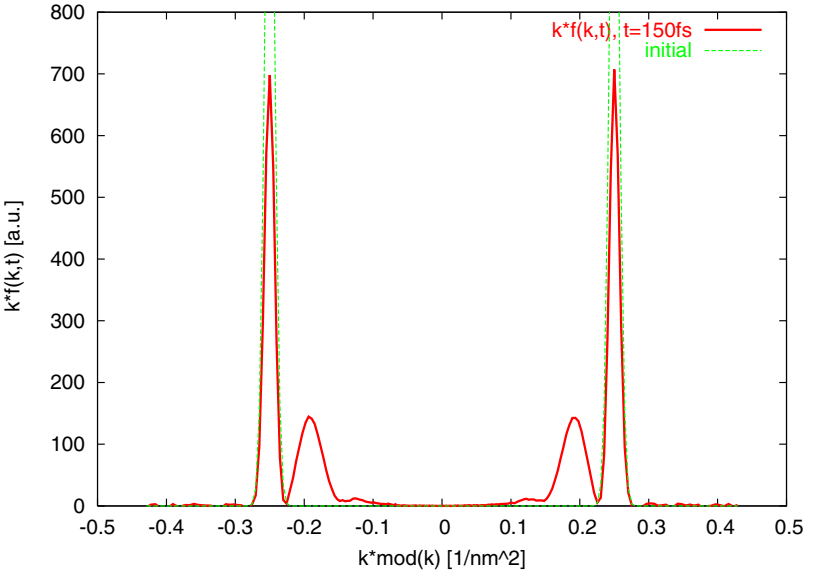

Fig. 2. Initial condition and energy distribution at $150fs$ evolution time

with respect to the initial condition. Also the place of the second replica can be recognized. In the absence of electric field there is a symmetry in the k_z directions. The behavior is analogous to the homogeneous case despite that the

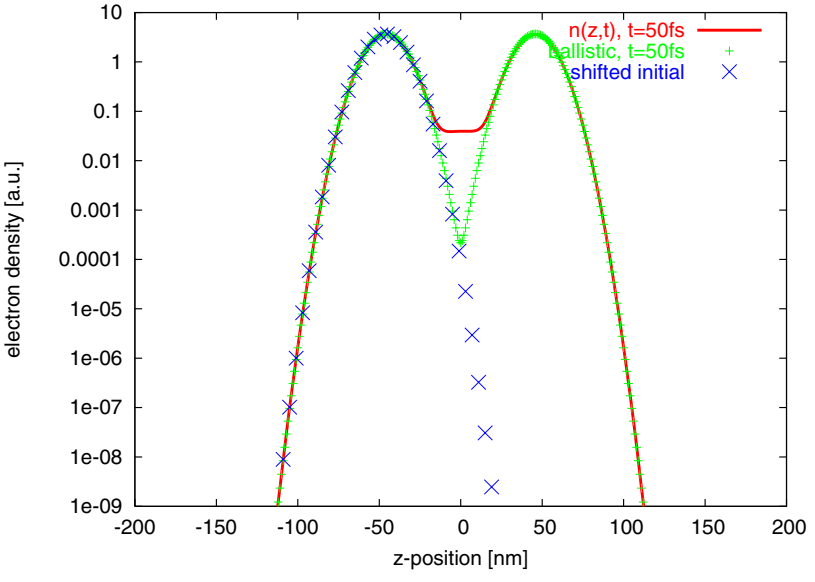

Fig. 3. Electron density after 50fs evolution time

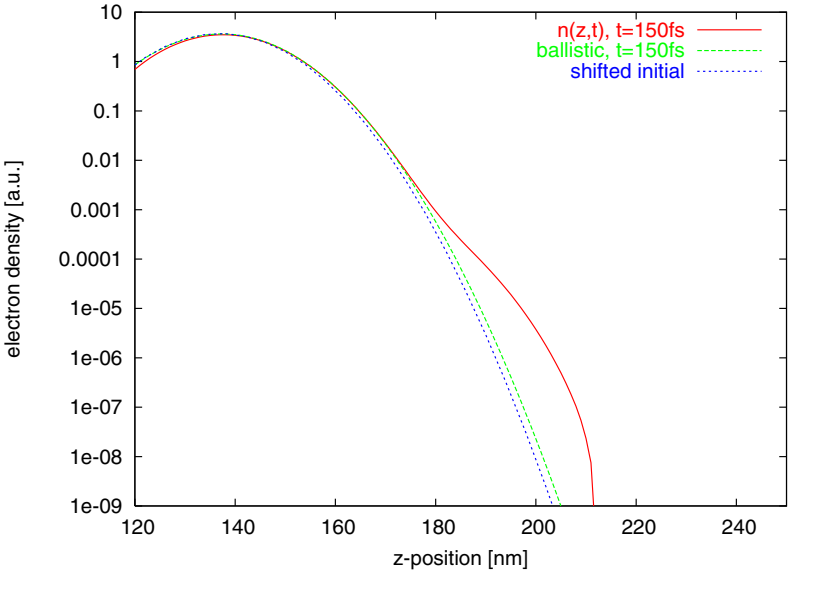

Fig. 4. Electron density after 150fs evolution time

coupling constant is now modified by G. Figure 3 compares the electron density with (n) and without (ballistic) electron-phonon interaction for 50 femtoseconds evolution time. The initial peak at the origin splits into two symmetric distri-

butions which evolve to the left and right respectively. In the central part the n curve is much higher than the ballistic curve due to the electrons which are slowed down by the phonons. The external fronts of the two curves coincide and hence are formed by the fastest electrons in the initial condition. The third curve is the shifted to the left half of the initial condition. It shows that at such small evolution time the real space broadening is practically zero. The same quantities (the half initial condition shifted to the right) are compared for 150 femtoseconds evolution time in Fig. 4. A window in the position is chosen for a better resolution. The broadening of the ballistic curve already becomes sensible. It shows the largest distance away from the origin which classical electrons can attain. The most interesting effect is demonstrated by the n curve: there is an excess electron density below approximately four orders of magnitude of the peak value. Such electrons penetrate in the semi-classically forbidden spatial zone. This purely quantum effect is due to the electrons, which occupy the energy region above the initial energy distribution. This effect has been recently observed in the solutions of a density matrix model of the zero field physical problem [3].

A Wigner equation for the evolution of spatially inhomogeneous electron distribution excited by a laser pulse in a quantum wire has been derived and solved by a Monte Carlo approach. A transformation is proposed which fixes the problem with the spreading integration domain in presence of electric field. It is shown that the quantum character of the electron-phonon interaction causes at low temperatures a speed-up effect on the electron front evolving in the wire. The proposed approach is suitable for exploration of the influence of the field on this effect. The numerical burden increases with the increase of the evolution time and requires large scale computational solutions such as parallel and GRID technologies.

Acknowledgments

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