Schrödinger's Equation as a Model Approach to Short Time-Scale Quantum Kinetics in a Semiconductor

J. A. Kenrow
Department of Physics, University of Florida, Gainesville, Florida 32611

T. K. Gustafson
Department of Electrical Engineering and Computer Science, University of California, Berkeley, California 94720
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We present a new approach to quantum kinetics based on a Schrödinger's equation formalism. As an example we apply this method to a coupled electron-LO phonon system in a semiconductor. We demonstrate that it is possible to compute the full many-body wave function of the electron-phonon system for short times. The time-dependent electron probability distribution, extracted from the many-body wave function, illustrates the non-Markovian nature of the early time kinetics. The retarded onset of dissipation and kinetic energy overshoots are explained through virtual, nonresonant transitions. [S0031-9007(96)01484-6]

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It is well known in high field transport theory for semiconductor microstructures [1–3] and for optically excited semiconductors using ultrashort laser pulses [4–6] that kinetic theories beyond the semiclassical Boltzmann kinetics are needed. In the Boltzmann picture, collisions are treated as being pointlike in space and time and individually conserve energy, while in the two cases mentioned above the carriers behave, at least to some extent, like quantum mechanical waves [7]. Appropriate quantum kinetic equations have been derived using reduced density matrices [4,8,9] and the Keldysh nonequilibrium Green's functions [5,6,10,11]. However, to obtain a closed set of kinetic equations for the one-particle expectation values, additional approximations are required. In the case of the reduced density matrices, one has to break the hierarchy of equations of motion and retain only the coupling to the next order correlation in some phenomenological (Markovian) manner [9]. In the case of nonequilibrium Green's functions, one has to choose an approximation for the self-energy and further use the generalized Kadanoff-Baym anzatz to reduce the two-timed, kinetic nonequilibrium Green's function to the one-timed density matrices. This anzatz requires a further assumption concerning the spectral nonequilibrium Green's functions [3,6].

The above approaches are well tested in the long time limit and in near equilibrium situations, both yielding Boltzmann-like kinetic equations. Quantum kinetic equations derived using these approaches have also been successfully applied to ultrashort time kinetics [4,8,9,12]. Unfortunately, the physical implications of the specific approximations for ultrashort time scales (especially in the case of nonequilibrium Green's functions) are not always well understood, and there are ongoing discussions to clarify this point [13,14]. Therefore, it is useful to have other independent formalisms, well suited for early time kinetics, which serve as models for comparing predictions obtained from the two kinetic theories mentioned. Several model systems have been recently proposed, i.e., the electron interaction with static disorder using the coherent potential approximation [15], and the electron-LO phonon interaction based on the Jaynes-Cummings model [16] and the Tomonaga-Luttinger model [17]. In this paper, we present a new approach to quantum kinetics based on the Schrödinger equation in which we develop a method to solve directly for the full many-body wave function for early times.

As an example we apply this theory to the short-time kinetics of carrier relaxation due to LO phonon emission. We consider the model system of a single quasi-1D conduction electron coupled to the complete spectrum of LO phonon modes in an ideal semiconductor quantum wire. The wire cross section ($\ell_x \times \ell_y$) lies in the x, y plane and the wire length along the z coordinate. Considering only intrasubband transitions we can take the full many-body wave function to be separable in the spatial coordinates, $|\Psi(\vec{r})\rangle = b(x, y)\,|g(z, t)\rangle$, where $b(x, y)$ is the lowest subband transversal eigenfunction of the wire [18]. We are then left with an effective 1D problem where the coupled electron-LO phonon system is represented by the longitudinal wave function, $|g(z, t)\rangle$, where the *ket* denotes the phonon state at time t given that the electron is at position z. In an interacting system, $|g(z, t)\rangle$ is not separable in the electron and phonon coordinates and it contains *all* the correlations (phase relations) between the electron and phonons. The many-body wave function, $|g(z, t)\rangle$, is time evolved within the effective mass approximation according to Schrödinger's equation,

$$i\hbar \frac{\partial |g\rangle}{\partial t} = \hat{H}_{tot}|g\rangle = (\hat{H}_e + \hat{H}_{\rm ph} + \hat{H}_{e-p})|g\rangle, \quad (1)$$

where $\hat{H}_e$ and $\hat{H}_{\rm ph}$ are the respective noninteracting electron and phonon Hamiltonian operators and $\hat{H}_{e-p}$ is the

effective 1D Fröhlich electron-phonon interaction Hamiltonian operator for the rectangular wire [19], all given, respectively, by

$$\hat{H}_e(z) = -\frac{\hbar^2}{2m}\frac{\partial^2}{\partial z^2}, \tag{2}$$

$$\hat{H}_{\rm ph} = \hbar\omega \sum_{\ell}^{N} (a_{q_\ell}^{\dagger} a_{q_\ell} + 1/2), \tag{3}$$

and

$$\hat{H}_{e-p}(z) = S \sum_{\ell}^{N} \frac{1}{Q_\ell} [a_{q_\ell} e^{iq_\ell z} - a_{q_\ell}^{\dagger} e^{-iq_\ell z}], \tag{4}$$

where N is the number of phonon modes, S is the effective 1D coupling constant, and $Q_\ell = [q_\ell^2 + (\pi/\ell_x)^2 + (\pi/\ell_y)^2]^{1/2}$. In Eqs. (2)–(4), m is the electron effective mass, and $a_{q_\ell}^{\dagger}(a_{q_\ell})$ denote the creation and (annihilation) operators for the LO phonons. The phonons are assumed to be dispersionless with a fixed energy of $\hbar\omega$. We define q_ℓ as the phonon wave number of the ℓth phonon mode corresponding to the z coordinate along the wire. We assume an uncorrelated initial state for the electron-phonon system which implies weak coupling. At $t = 0$, the electron is given by a Gaussian wave packet along z with specified wave number k_i. The lattice at $t = 0$ is taken as the vacuum state for the LO phonons.

A solution to Eq. (1) is obtained using a Schrödinger representation in which the time harmonic dependence for both the lattice mode vibrations and electron wave packet are contained in $|g(z,t)\rangle$. We construct $|g(z,t)\rangle$ by first considering the time evolution of the phonon states alone,

$$i\hbar \frac{\partial |g(t)\rangle}{\partial t} = \hat{H}_{\rm ph} |g(t)\rangle. \tag{5}$$

The solution for the phonon wave function, $|g(t)\rangle$, according to Eq. (5) can be written as a linear superposition over the orthonormal basis of LO phonon number states,

$$|g(t)\rangle = \alpha(t)\,|0\rangle + \sum_{\ell}^{N} \beta_\ell(t) e^{-i\omega t} |1\rangle_\ell + \sum_{\ell,\, m\geq\ell}^{N} \gamma_{\ell m}(t) e^{-i2\omega t} |2\rangle_{\ell m} + \ldots, \tag{6}$$

where $|0\rangle$ represents the lattice vacuum state, $|1\rangle_\ell$ and $|2\rangle_{\ell m}$ represent, respectively, first- and second-order number states, and the coefficients α, β_ℓ, and $\gamma_{\ell m}$ are the corresponding weighting factors for each lattice state at time t. In Eq. (6) the energies are counted from the lattice zero point energy for convenience.

For the purpose of explicitly illustrating the ordering of the phonon mode occupancies in the various number states, we let $|1\rangle_\ell$ represent the distinct first-order number state with an occupancy of **1** in the ℓth LO phonon mode. All the possible second-order number states, $|2\rangle_{\ell m}$, span the $\ell \times m$ matrix which for $\ell = m$, $|2\rangle_{\ell\ell}$ represents the lattice state with an occupancy of **2** in the ℓth LO phonon mode and for $\ell \neq m$, $|2\rangle_{\ell m}$ represents the lattice state with an occupancy of **1** in both the ℓth and mth LO phonon modes, e.g.,

$$\begin{aligned} |0\rangle &= |000\cdots0\rangle \qquad \text{(vacuum state)} \\ |1\rangle_\ell &= |0\cdots\tfrac{1}{\ell}\cdots0\rangle \qquad \text{1st order,} \\ |2\rangle_{\ell m} &= |0\cdots\tfrac{1}{\ell}\cdots\tfrac{1}{m}\cdots0\rangle \quad m > \ell \quad \text{2nd order,} \\ |2\rangle_{\ell m} &= |0\cdots\tfrac{2}{\ell}\cdots0\rangle \quad \ell = m \quad \text{2nd order.} \end{aligned}$$

To avoid duplicate counting and to ensure distinct second-order states, it is required that $m \geq \ell$. It follows that for a system of N phonon modes, there are N distinct combinations of first-order number states, $|1\rangle_\ell$, and $N(N+1)/2$ distinct combinations of second-order number states, $|2\rangle_{\ell m}$.

The complete coupled electron-phonon solution, $|g(z,t)\rangle$, is obtained by inserting back in the electron coordinate,

$$|g(z,t)\rangle = \alpha(z,t)\,|0\rangle + \sum_{\ell}^{N} \beta_\ell(z,t) e^{-i\omega t} |1\rangle_\ell + \sum_{\ell,\, m\geq\ell}^{N} \gamma_{\ell m}(z,t) e^{-i2\omega t} |2\rangle_{\ell m} + \cdots, \tag{7}$$

where all the electronic information is contained in and extracted from the coefficients α, β_ℓ, and $\gamma_{\ell m}$. The final set of coupled kinetic equations for $\alpha(z,t)$, $\beta_\ell(z,t)$, and $\gamma_{\ell m}(z,t)$ are obtained by applying the Hamiltonian operators defined by Eqs. (2)–(4) as well as $\hat{H}'(z)$ on $|g(z,t)\rangle$ and then projecting onto each unique number state

$$\frac{\partial \alpha}{\partial t} = \frac{1}{i\hbar}\left[-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial z^2}\right]\alpha + \frac{S e^{-i\omega t}}{i\hbar} \sum_{\ell}^{N} \frac{e^{iq_\ell z}}{Q_\ell}\beta_\ell, \tag{8}$$

$$\frac{\partial \beta_\ell}{\partial t} = \frac{1}{i\hbar}\left[-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial z^2}\right]\beta_\ell - \frac{S e^{i(\omega t - q_\ell z)}}{i\hbar Q_\ell}\alpha + \frac{S e^{-i\omega t}}{i\hbar} \times \left[\frac{e^{iq_\ell z}}{Q_\ell}\sqrt{2}\,\gamma_{\ell\ell} + \left(\sum_{\substack{m \\ m<\ell}}^{N} \gamma_{m\ell} + \sum_{\substack{m \\ m>\ell}}^{N} \gamma_{\ell m}\right)\frac{e^{iq_m z}}{Q_m}\right], \tag{9}$$

$$\frac{\partial \gamma_{\ell m}}{\partial t} = \frac{1}{i\hbar}\left[-\frac{\hbar^2}{2m}\frac{\partial^2}{\partial z^2}\right]\gamma_{\ell m} - \frac{\sqrt{2}\,S e^{i(\omega t - q_\ell z)}}{i\hbar Q_\ell}\beta_\ell \delta_{\ell m} - \frac{S e^{i\omega t}}{i\hbar}\left[\frac{e^{-iq_\ell z}}{Q_\ell}\beta_m + \frac{e^{-iq_m z}}{Q_m}\beta_\ell\right]\bar{\delta}_{\ell m} + \cdots, \tag{10}$$

where $\delta_{\ell m}$ is the Kronecker delta function and we define $\bar{\delta}_{\ell m} \equiv 1$ (for $\ell \neq m$) as the "anti" Kronecker delta function.

The set of Eqs. (8)–(10) has complete time reversal symmetry. In contrast to Boltzmann kinetics, where all phase coherence is destroyed after one single scattering

event, the phase coherence here is *never* lost but instead distributed over a large number of phonon modes. Therefore, the presented quantum kinetic model bridges between the Boltzmann picture and the quantum kinetic model based on the Jaynes-Cummings model [16], where the phase can only oscillate between a two-level atom and a single phonon mode.

From the set of Eqs. (8)–(10) we observe that at $t = 0$, only the coefficient α is nonzero and at small finite times $\beta_\ell \sim \kappa t$ and $\gamma_{\ell m} \sim (\kappa t)^2$, where $\kappa = (S/\hbar Q_\ell)|_{q_\ell = 0}$ is the maximum coupling strength. The next higher coefficients, i.e., for the three phonon state, are of order $\mathcal{O}[(\kappa t)^3]$. This reflects the fact that in Eq. (4) the electron is coupled to one phonon at a time. In higher order phonon processes the electron must emit or absorb one phonon first, before it can emit or absorb the next one. In contrast to the Boltzmann picture, the emission or absorption of the first phonon is not necessarily complete, i.e., the intermediate electron state can be virtual. Nevertheless, coupling to higher order number states is retarded by a time $t \sim 1/\kappa$. In the case considered here, this retardation time is 400 fs. It follows that for small coupling strengths and small times the trajectory of the full many-body system is confined to the subspace of **0**, **1**, and **2** phonon number states. We can make use of this fact by replacing the full Hamiltonian in Eq. (1) by its projection onto the subspace of **0**, **1**, and **2** phonon number states, $\hat{H}_{tot} \rightarrow P\hat{H}_{tot}P$, where P is the projection operator defined by $P|0\rangle = |0\rangle$, $P|1\rangle_\ell = |1\rangle_\ell$, $P|2\rangle_{\ell m} = |2\rangle_{\ell m}$, and $P|n\rangle = 0$ for $n \geq 3$. In this subspace the problem is exactly solvable. We note that this projection onto a finite number of degrees of freedom leads to a closed system where the kinetics can never truly be dissipative. On very short times, however, the kinetics of this closed system are indistinguishable from true dissipative kinetics with infinite degrees of freedom.

Equations (8)–(10) are solved to obtain the wave function $|g(z,t)\rangle$ of the coupled electron-phonon system and hence the total probability density $\rho(z,t) = |g(z,t)\rangle\langle g(z,t)|$. The electronic probability density is then formed by taking a partial trace over the lattice coordinates,

$$\begin{aligned}\rho_e(z,t) &= \mathrm{Tr}_L[\rho(z,t)] \\ &= |\alpha(z,t)|^2 + \sum_{\ell}^{N} |\beta_\ell(z,t)|^2 \\ &\quad + \sum_{\ell,\, m \geq \ell}^{N} |\gamma_{\ell m}(z,t)|^2. \end{aligned} \tag{11}$$

We illustrate the short-time kinetics by numerical integration of the projected Eqs. (8)–(10). For the GaAs wire we used material parameters ($m = 0.067 m_e$, $\hbar\omega = 36.2$ meV, and $\ell_x = \ell_y = 60$ Å). The constraint of the initial lattice vacuum state gives rise to $\beta_\ell(z,0) = \gamma_{\ell m}(z,0) = 0$ for all m and ℓ such that $|g(z,0)\rangle = \alpha(z,0)\,|0\rangle = \exp\{-0.5[(z - z_o)/\Delta z_i]^2 + ik_i z\}\,|0\rangle$ is just the initial Gaussian centered at $z = z_o$ with spread Δz_i and initial wave number k_i.

In the following numerical studies, the energy and wave number have been scaled, respectively, by, $E_o \equiv \hbar\omega = 36.2$ meV and $a_o \equiv \hbar/\sqrt{2mE_o} = 3.975$ nm. In Fig. 1 the average electron wave number $\langle k\rangle$ and kinetic energy $\langle \epsilon_k\rangle = \langle \hbar^2 k^2/2m\rangle$ are shown as a function of time for initial energy $E_i = 0.1$ eV. The initial Gaussian spread was chosen to be $\Delta k_i a_o = 0.2$. The non-Markovian behavior of the kinetics can be seen clearly in both sets of curves from the following features. In Fig. 1(a) the dissipative behavior of the electron wave number clearly deviates from the well known semiclassical (exponential) decay, illustrated by the dashed line. In fact, for the first few femtoseconds there is virtually no dissipation, the electron "appears" to propagate freely along the wire undisturbed by the lattice. Only until times, $t \sim 1/\omega$ (inverse phonon frequency), does the electron begin to "feel" the lattice and dissipate through phonon emission. Another feature is illustrated in Fig. 1(b) where in the first few femtoseconds, the electron exhibits a slight increase in energy before dissipation begins. This seemingly unphysical behavior can be understood from the electron probability distribution, shown in Fig. 2 for times $t = 0, 20, 50$, and 100 fs. The curve for $t = 20$ fs illustrates a large, nearly structureless spread, in the final electron state and only until later times does a reshaping begin to appear at the positions of the first phonon replicas centered at $ka_o = \pm 1.33$ (the second phonon replicas at $ka_o = \pm 0.83$ are barely noticeable). This large spread in the distribution is a direct consequence of the time-energy uncertainty. Classically, the final electron state, ϵ_{k-q}, is determined by selection rules governed by the energy conserving relation, $|\epsilon_k - \epsilon_{k-q} - \hbar\omega| = 0$. However, for small finite times, an energy uncertainty $\Delta\epsilon$ exists, allowing virtual transitions to the final state for times $\Delta t = \hbar/\Delta\epsilon$ governed by the relation $|\epsilon_k - \epsilon_{k-q} - \hbar\omega| = \Delta\epsilon$. These virtual transitions produce an

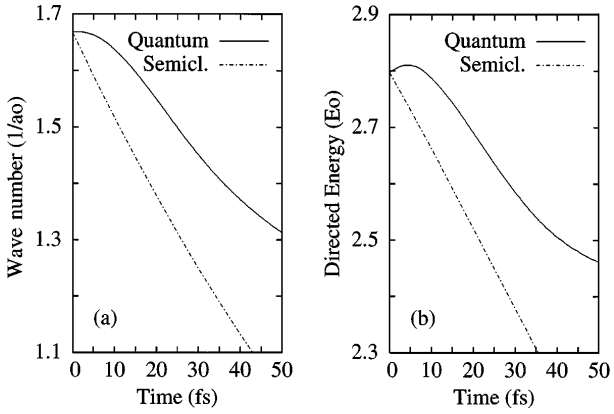

FIG. 1. (a) The average electron wave number $\langle k\rangle$, and (b) the kinetic energy $\langle \epsilon_k\rangle$ as a function of time for initial electron energy $E_i = 0.10$ eV. The corresponding semiclassical result is shown for comparison.

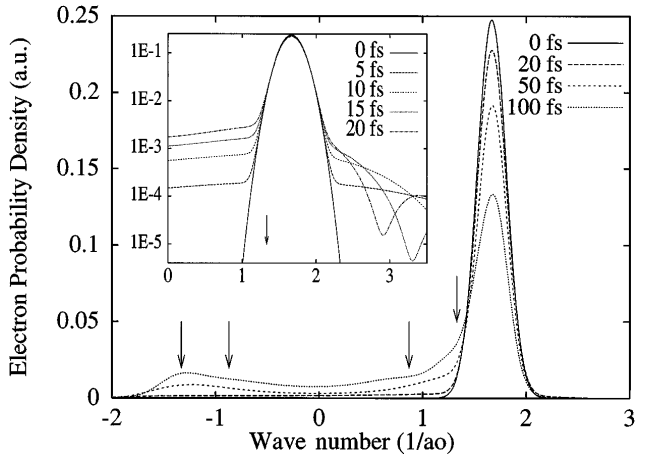

FIG. 2. The electron probability distribution $\rho_e(k,t)$ as a function of wave number k with $E_i = 0.1$ eV at times $t = 0, 20, 50$, and 100 fs. The arrows mark the center positions of the forward and backscattering first and second phonon replicas. The inset is a blowup of the first 20 fs ($t = 0, 5, 10, 15$, and 20 fs) of $\rho_e(k,t)$ where the arrow marks the forward scattering first phonon replica.

essentially symmetrically broadened final electron state. A blowup of the electron distribution for times $t \leq 20$ fs is shown in the inset of Fig. 2. For times $t \ll 1/\omega$ the energy spread is so large that the electron has almost equal probability for gaining energy versus losing energy (see inset for $t = 5$ fs). This wide symmetrical spread leaves $\langle k \rangle$ unchanged but leads to an increase in $\langle \epsilon_k \rangle$. As time increases, however, the allowed transitions (centered around the first phonon replica at $ka_o = 1.33$) increase in amplitude while the forbidden transitions at higher k values die out in an oscillatory manner. Thus for times $t \sim 1/\omega$ the distribution of final states is already so narrow that the electron loses energy through each transition and an onset of dissipation can be seen. The kinetic energy overshoot can also be explained in terms of a correlation buildup. At early times the electron is losing potential energy through a deformation of the lattice, which leads to an increase in its kinetic energy. Similar behavior has been presented by Schilp *et al.* [9] in which they derived quantum kinetic equations using reduced density matrices for single band electron dissipation in the presence of LO phonons in 3D bulk GaAs.

In conclusion, we have presented a new approach to quantum kinetics based on a Schrödinger equation formalism. It was shown that for early times only a small portion of the many-body Hilbert space is involved in the evolution of the electron-LO phonon system. We demonstrated that by restricting the Hamiltonian to a finite subspace, the full many-body wave function can be obtained without any approximations. This wave function approach thus accounts for *all* the many-body correlations in the finite subspace. The early time kinetics were demonstrated through temporal studies of the average electron wave number and kinetic energy. For early times, a retardation in the electron-phonon interaction was explained through a buildup of correlation which is a direct consequence of the time-energy uncertainty. Because of its pure quantum mechanical nature, the presented wave function method gives a very intuitive approach to quantum kinetics.

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