A GENERALIZED MONTE CARLO APPROACH FOR THE SIMULATION OF THE COHERENT ULTRAFAST DYNAMICS IN PHOTOEXCITED SEMICONDUCTORS

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Abstract

In this paper a generalized Monte Carlo method recently developed by the authors for the solution of the coupled set of quantum kinetic equations for the distribution functions and the interband polarization is presented. The aim of this method is to combine the advantages of the description within a fully quantum mechanical picture with the power of the Monte Carlo technique for the treatment of stochastic processes. It is based on a decomposition of the kinetic equations in a coherent and an incoherent part. The former is integrated directly while the latter is sampled by means of a Monte Carlo simulation. This allows us to treat on the same kinetic level carrier thermalization and relaxation as well as dephasing processes.

I. INTRODUCTION

The Monte Carlo (MC) method, which has been applied for more than 25 years to the analysis of semiclassical charge transport in semiconductors, is the most powerful numerical tool for microelectronic device simulation [1]. On the other hand, the present-day technology allows the investigation of relaxation and dephasing phenomena in semiconductors with a time resolution which has now reached a few femtoseconds [2]. On such a time-scale, coherent aspects play an important role and the carrier dynamics cannot be treated in terms of the traditional semiclassical transport theory. Therefore, in order to study this partially coherent dynamics, a generalization of the conventional MC method is required.

The aim of the present invited paper is to review a method recently proposed by the authors [3] and to discuss its application to the analysis of ultrafast carrier dynamics in photoexcited semiconductors [4, 5]. The main peculiarity of the method is to retain the big advantages of the MC method in treating scattering processes and, at the same time, to take into account on the same kinetic level also coherent phenomena. Compared to the conventional MC technique, which simply provides a solution of the semiclassical Boltzmann Transport Equation (BTE), this generalized MC approach provides a solution of the Semiconductor Bloch Equations (SBE). In addition to a simulation of the various distribution functions, this will result in a simulation of the interband polarization induced by the coherent light field.

Such an approach allows a selfconsistent description of the carrier photogeneration process [5]. The energy broadening due to the finite pulse duration and due to the decay of the interband polarization has not to be introduced as a phenomenological parameter as in any conventional MC simulation [6] but it comes out selfconsistently with its full time dependence.

II. PHYSICAL SYSTEM AND THEORETICAL APPROACH

Let us consider a bulk-semiconductor model characterized by two spherical and parabolic bands. In a semiclassical picture, only the carrier distribution functions over single-particle states are considered as kinetic variables. All interactions between carriers and other types of quasiparticles, and in particular also the interaction with the external light field, are treated as perturbations. They are usually described in terms of scattering processes within Fermi's Golden Rule. Such approximations lead to the BTE for the distribution functions of electrons ($f^e_{\mathbf{k}}$) and holes ($f^h_{\mathbf{k}}$):

$$\frac{d}{dt} f^{e,h}_{\mathbf{k}} = \sum_{\mathbf{k}'} [s^{e,h}_{\mathbf{k},\mathbf{k}'} f^{e,h}_{\mathbf{k}'} - s^{e,h}_{\mathbf{k}',\mathbf{k}} f^{e,h}_{\mathbf{k}}] \tag{1}$$

with scattering rates $s^{e,h}_{\mathbf{k}',\mathbf{k}}$. On this level all coherence or correlation effects are neglected.

For the analysis of coherent phenomena, the phase relation between different types of carriers, induced by the light-matter interaction, has to be treated explicitly [3, 7, 8]. Therefore, the kinetics cannot be simply described in terms of distribution functions (intraband density matrices) but we have to include as kinetic variables also the interband polarization (interband density matrix) $p_{\mathbf{k}}$. For the unperturbed dynamics a closed set of equations can be easily obtained [3]. However, this fully coherent dynamics is modified by the presence of the various interaction mechanisms. They give rise to an infinite hierarchy of equations of motion which has to be truncated at some level.

Here, we will limit ourselves to contributions up to second order in the interaction matrix elements. The second-order terms are treated within the usual Markov approximation [3] and all second-order contributions which involve second or higher powers of the polarization are neglected. Within such an approximation scheme, the resulting system of SBE takes the general form [3]:

$$\frac{d}{dt} f^e_{\mathbf{k}} = g_{\mathbf{k}}(t) + \sum_j \frac{d}{dt} f^e_{\mathbf{k}}\Big|^j_{inco}, \qquad \frac{d}{dt} f^h_{\mathbf{k}} = g_{-\mathbf{k}}(t) + \sum_j \frac{d}{dt} f^h_{\mathbf{k}}\Big|^j_{inco}, \tag{2}$$

$$\frac{d}{dt} p_{\mathbf{k}} = \frac{1}{i\hbar}\Big\{(\mathcal{E}^e_{\mathbf{k}} + \mathcal{E}^h_{-\mathbf{k}} + \Omega_{\mathbf{k}}) p_{\mathbf{k}} + (M_{\mathbf{k}} A_0(t) e^{-i\omega_L t} + \Delta_{\mathbf{k}})(1 - f^e_{\mathbf{k}} - f^h_{-\mathbf{k}})\Big\} + \sum_j \frac{d}{dt} p_{\mathbf{k}}\Big|^j_{inco}, \tag{3}$$

with a generation rate

$$g_{\mathbf{k}} = \frac{1}{i\hbar}[(M_{\mathbf{k}} A_0(t) e^{-i\omega_L t} + \Delta_{\mathbf{k}}) p^*_{\mathbf{k}} - (M^*_{\mathbf{k}} A^*_0(t) e^{i\omega_L t} + \Delta^*_{\mathbf{k}}) p_{\mathbf{k}}], \tag{4}$$

where $A_0(t)$ is the envelope of the vector potential of the external light field with angular frequency ω_L, $M_{\mathbf{k}}$ is the dipole matrix element, and the index j refers to the various interaction mechanisms. They result to modify the system dynamics with two different contributions: (i) Coherent terms which lead to a renormalization of the free-carrier energies $\mathcal{E}^{e,h}_{\mathbf{k}}$ by a self-energy $\Omega_{\mathbf{k}}$ and of the external light field by an internal field $\Delta_{\mathbf{k}}$, and (ii) incoherent terms which lead to relaxation and dephasing processes.

Denoting by $\mathcal{F}_{\mathbf{k}}$ the generic kinetic variable (distribution functions or polarization), the SBE (2,3) can be schematically rewritten as

$$\frac{d}{dt}\mathcal{F}_{\mathbf{k}} = \frac{d}{dt}\mathcal{F}_{\mathbf{k}}\Big|_{co} + \frac{d}{dt}\mathcal{F}_{\mathbf{k}}\Big|_{inco}, \tag{5}$$

with a coherent part

$$\frac{d}{dt}\mathcal{F}_{\mathbf{k}}\Big|_{co} = C^0_{\mathbf{k}}(\{\mathcal{F}\}) + \sum_j C^j_{\mathbf{k}}(\{\mathcal{F}\}), \tag{6}$$

where $C^j_{\mathbf{k}}$ is some functional of the kinetic variables. Within the approximations discussed it is easy to realize a strong formal similarity between the various kinetic equations. In particular, the incoherent contributions

have exactly the same formal structure of the "Boltzmann collision term" in Eq. (1) also for the case of the interband polarization:

$$\frac{d}{dt}\mathcal{F}_{\mathbf{k}}\Big|_{inco} = \sum_{j}\sum_{\mathbf{k}'}[\mathcal{S}^{j}_{\mathbf{k},\mathbf{k}'}\mathcal{F}_{\mathbf{k}'} - \mathcal{S}^{j}_{\mathbf{k}',\mathbf{k}}\mathcal{F}_{\mathbf{k}}] \,, \tag{7}$$

where $\mathcal{S}^{j}_{\mathbf{k},\mathbf{k}'}$ denotes the scattering rate for a transition $\mathbf{k}' \rightarrow \mathbf{k}$ induced by the j-th interaction mechanism. This strong similarity constitutes the starting point of our generalized MC approach.

III. GENERALIZED MONTE CARLO PROCEDURE

As a starting point, let us briefly recall the basic ideas of the conventional MC simulation. As discussed above, the semiclassical transport theory is based on the BTE (1). This is, in general, a non-linear equation which is usually transformed into a locally linear one by means of a time-step solution. Due to this local linearity, the distribution function at any time t within the time-step can be written as

$$f^{e,h}_{\mathbf{k}}(t) = \sum_{\mathbf{k}'} G^{e,h}_{\mathbf{k},\mathbf{k}'}(t,t_o) f^{e,h}_{\mathbf{k}'}(t_o) \,, \tag{8}$$

where G, called Boltzmann propagator, has a direct physical interpretation: it describes the probability that a particle in state $\mathbf{k}'$ at time t_o will be found in state $\mathbf{k}$ at time t. Equation (1) and (8) can be regarded as the starting point of the traditional Ensemble Monte Carlo (EMC) technique [1, 11] which simply provides a MC sampling of the sum in Eq. (8). Such sampling is performed through a stochastic simulation of a suitable ensemble of carriers. These "simulative carriers" are, in general, not real physical particles; such an ensemble of particles is only representative for the real carrier system. For each simulative carrier, a sequence of random "free flights", interrupted by random "scattering events", is generated. It can be shown, that such a "random walk" in k-space is just a MC sampling of the Boltzmann propagator $G_{\mathbf{k}_f,\mathbf{k}_i}(t,t_o)$, where $\mathbf{k}_i$ and $\mathbf{k}_f$ denote, respectively, the initial and the final state of the generic random walk [11].

Let us now come back to the system of quantum kinetic equations (5). Since this is again a system of nonlinear equations, as in the semiclassical case, we introduce a time discretization. In the proposed numerical procedure, for each time step Δt, the coherent contributions (6) are evaluated by means of a direct numerical integration while the incoherent contributions (7) are "sampled" by means of a generalized MC simulation. Let us now focus our attention on the explicit form of the incoherent contributions (7): For all the kinetic variables (including the polarization field) the various $\mathcal{S}_{\mathbf{k},\mathbf{k}'}$ are within our approximations positive-definite quantities, i.e. they can be regarded as "true" scattering probabilities from state $\mathbf{k}'$ to state $\mathbf{k}$. However, the function $\mathcal{F}$ is now a complex function. Due to the local linearity of our transport equation over the time-step, the kinetic variable at time t can be written as

$$\mathcal{F}_{\mathbf{k}}(t) = \sum_{\mathbf{k}'} \mathcal{G}_{\mathbf{k},\mathbf{k}'}(t,t_o)\mathcal{F}_{\mathbf{k}'}(t_o) \,, \tag{9}$$

where $\mathcal{G}$ is now a generalized Boltzmann propagator corresponding to the kinetic variable. As for the semiclassical case, the propagator $\mathcal{G}$ results to be a positive-definite solution of the generalized Boltzmann equation (7). Therefore, it can be again sampled by means of a conventional EMC simulation.

Equation (9) constitutes the starting point of our generalized MC approach. As for the semiclassical case, such sampling is again performed through a stochastic simulation of a suitable ensemble of carriers which, in general, have nothing to do with real physical particles. The structure of the proposed MC procedure can then be summarized as follows: The total time is divided into time-steps. The simulation starts before the laser has been switched on. The system is chosen to be in its fundamental state, i.e. the vacuum of electron-hole pairs. The simulation then results in a loop over the various time steps. For each time step:

(i) we introduce an ensemble of $N_{\mathbf{k}}(t_o)$ "simulative particles" with $N_{\mathbf{k}}(t_o) \propto |\mathcal{F}_{\mathbf{k}}(t_o)|$; (ii) we attach to each "particle" i a phase-factor w_i according to the phase of $\mathcal{F}_{\mathbf{k}}(t_o)$; (iii) for each "particle" we sample its propagator $\mathcal{G}$ by means of a conventional EMC simulation [1] i.e. a random sequence of free flights and scattering events; (iv) at the end of the time-step the new value of $\mathcal{F}$ is evaluated: $\mathcal{F}_{\mathbf{k}}(t_o + \Delta t) = \sum_i w_i$. The usual "counting" of the particles in k is then replaced by a sum of these phase-factors w_i which reflects the complex nature of the kinetic variable $\mathcal{F}$. A similar approach has been recently used by the authors for a MC simulation of four-wave mixing experiments [4]. In this case, a MC simulation of the various, in general complex, Fourier components of the distribution functions is required.

IV. APPLICATIONS

We will now present some numerical results concerning simulations characterized by a laser energy far from the band gap (excess energy $\mathcal{E}_{ex} = 0.18$ eV, pulse duration $\tau_L = 50$ fs). This is the typical situation for energy-relaxation experiments [2].

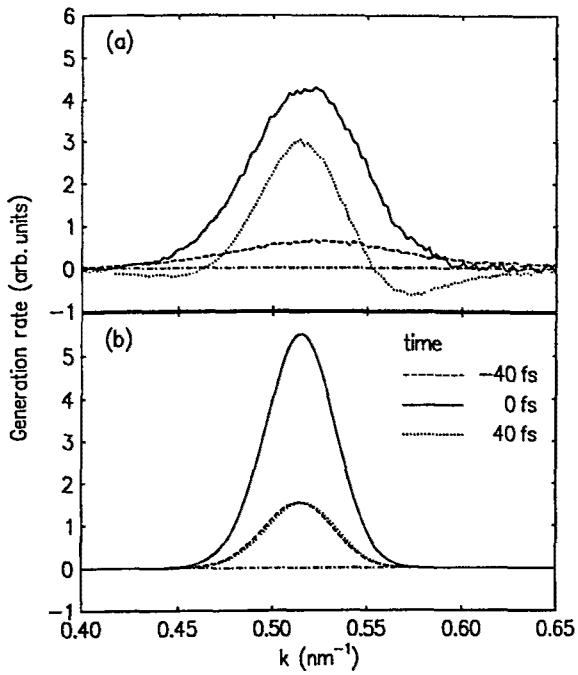

FIG. 1. Generation rates for a final density $n = 10^{16}$ cm^{-3}, (a) obtained from the SBE, and (b) in the semiclassical limit.

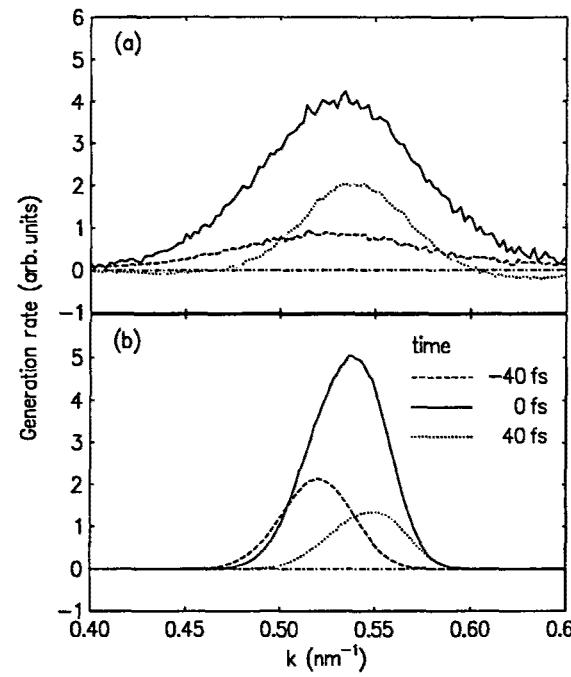

FIG. 2. Same as FIG. 1, but for a final density $n = 10^{18}$ cm^{-3}.

For a better understanding, let us first consider a "simulated experiment" characterized by a final carrier density $n = 10^{16}$ cm^{-3}. In Fig. 1 the self-consistent generation rates obtained from this MC simulation are shown as a function of the wave-vector k for different times during the laser pulse. Figure 1 (a) shows the generation rates for the full generation model while in (b) the corresponding rates for the semiclassical case are plotted. Due to the Markovian limit, the latter ones do not contain regions with negative values. On the contrary, the rates in (a) exhibit a strong time-dependence also in the shape. In particular, at short times, the shape of the generation rate is found to be much broader than estimated from the uncertainty principle using the pulse width as uncertainty of time. The reason is that the "observation time" has to be used for a correct

estimation of the line width. For longer times we note a narrowing of the generation rate, this narrowing, however, is accompanied by the build-up of negative regions off-resonance which can be interpreted as a stimulated recombination process. Thus, the distribution of the generated carriers does not only become narrower with increasing time due to a generation mainly in resonance but also due to a recombination of those carriers which have been generated performing "energy non-conserving transitions" at short times.

The above result shows that a self-consistent treatment of the generation process can be important if either the evolution is analyzed already during the pulse or if some scattering mechanism is so strong that it can remove those carriers generated with the "wrong" energy before they can recombine. The latter one is exactly the situation that we obtain by repeating the above "simulated experiment" for the case of a final density $n = 10^{18}\,\mathrm{cm}^{-3}$. The self-consistent generation rates obtained in this case are shown in Fig. 2. Due to the strong efficiency of carrier-carrier scattering, already during the laser pulse carriers are removed from their initial distribution and, therefore, the stimulated recombination of Fig. 1(a) is strongly reduced. As a consequence, in this case the carrier distribution after the end of the laser pulse results to be significantly broader compared to the corresponding semiclassical case. The energy shift of the generation rate especially in the high density case is due to band gap renormalization. This effect is present in the semiclassical model as well as in the full generation model. Phase space filling effects lead to the asymmetries in the generation rate in the high density case at 0 fs and especially at 40 fs.

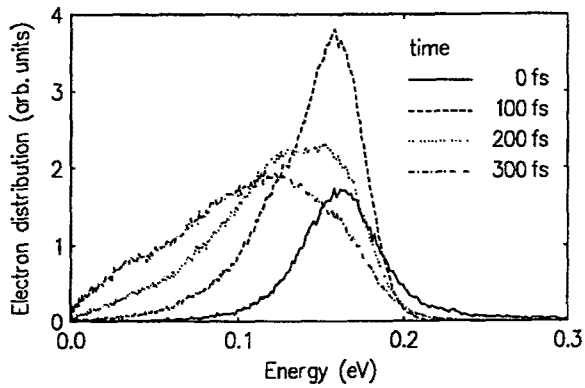

FIG. 3. Electron energy distribution for different times.

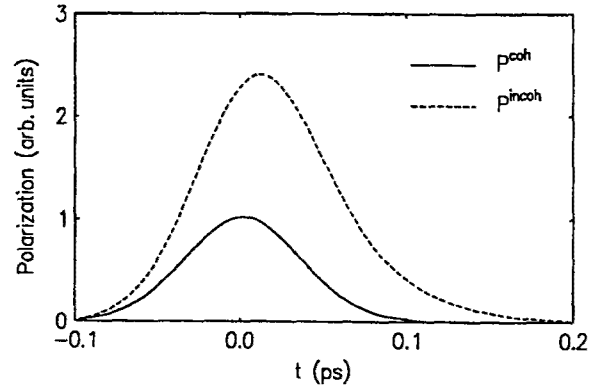

FIG. 4. Polarization as function of time.

In Fig. 3 the electron distribution as a function of energy at different times is also shown. It corresponds to the first simulated experiment ($n = 10^{16}\,\mathrm{cm}^{-3}$) with the full generation model (see Fig. 1(a)). The figure shows the typical scenario of carrier thermalization due to carrier-carrier scattering and energy relaxation due to carrier-phonon scattering. In the electron energy distribution we notice still some structure related to the discrete emission of optical phonons, which becomes more pronounced in the case of lower densities.

In Fig. 4 the polarization of the carrier system is shown as a function of time: The solid curve refers to the absolute value of the total polarization $P^{coh} = |\sum_{\mathbf{k}} p_{\mathbf{k}}|$. It decays due to the inhomogeneous broadening in k-space since each contribution $p_{\mathbf{k}}$ in the sum rotates with a different frequency. Its decay strongly depends on the width of the carrier distribution and, therefore, on the properties of the laser pulse, and its time scale is typically much shorter than that related to incoherent phenomena. The dashed curve refers to the incoherently summed polarization $P^{incoh} = \sum_{\mathbf{k}} |p_{\mathbf{k}}|$. It is a measure of the degree of coherence still present in the system and after the pulse it decays due to incoherent scattering processes. The dephasing time is of the order of 100 fs which in this case is mainly due to carrier-carrier interaction.

V. CONCLUSIONS

We have presented a numerical method which enabled us to include coherent phenomena in a MC simulation. The theory is based on the SBE for the distribution functions of electrons and holes, as well as for the interband polarization. Within this method the generation process is treated in a self-consistent way with its full time-dependence. We have obtained the time-dependence of the total polarization as well as of the incoherently summed polarization, which describes the degree of coherence still present in the system. Thus we can analyze the various times relevant for the dephasing process.

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