Analysis of Coherent and Incoherent Phenomena in Photoexcited Semiconductors: A Monte Carlo Approach

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A generalized Monte Carlo procedure for the ultrafast dynamics of photoexcited carriers in a semiconductor is presented, where the coherence in the carrier system as well as band renormalization and excitonic effects in the Hartree-Fock approximation are fully taken into account. The details of the coherent generation process, the energy relaxation, and dephasing of the carriers are analyzed. The approach presents a numerical method for the investigation of phenomena occurring close to the band gap and those typical for the relaxation of hot carriers.

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The relaxation process of photoexcited carriers has been investigated with an increase in time resolution which has now reached a few femtoseconds [1-5]. The experimental results have been analyzed within continuously refined theoretical models. Most quantitative calculations have made use of Monte Carlo simulations [4,6-8], since this technique seems to be best suited to include a variety of interaction mechanisms on a microscopic level. In a Monte Carlo simulation the numerical efficiency is improved by performing an "important sampling" where the regions in k space are weighted by the distribution function. In all these simulations, scattering processes as well as the generation of carriers are treated as purely stochastic processes. Thus, the variety of coherent phenomena [9-12], which have attracted great interest during recent years, cannot be treated within these simulations. In this case, the theoretical analysis has to start from quantum kinetic equations [13-17] which, besides describing the carrier distribution functions, also contain information on the interband polarization.

In particular on short time scales the two classes of phenomena cannot be separated. The coherent light of an ultrashort laser pulse introduces coherence in the carrier system. If the physical phenomena under investigation occur on a time scale shorter than the "dephasing time," they should be taken into account in a theoretical analysis even if the measured quantities are variables purely determined by distribution functions. The aim of this Letter is therefore to present a method which retains the advantages of the Monte Carlo method in treating scattering processes in an efficient way and at the same time is capable of taking into account coherent effects.

The theoretical basis for the investigation of coherent phenomena in a photoexcited semiconductor is the generalized optical Bloch equations [13-17] for the distribution functions of electrons and holes and for the interband polarization. We consider a simplified two-band bulk GaAs model. The external light field is treated classically and within the rotating-wave approximation. A Gaussian pulse shape has been chosen. The carrier-carrier interaction is included in the screened Hartree-Fock approximation with a Lindhard-like static screening which is calculated self-consistently. It gives rise to a self-energy and an internal field (interband self-energy). The carrier-phonon interaction is included for both the polar optical and acoustic deformation potential coupling within the second-order Markov ("golden rule") approximation, leading to the standard electron-phonon self-energy and Boltzmann scattering terms.

The main features of this quantum kinetic treatment, which cannot be included in a traditional Monte Carlo simulation, are the phase relations between different types of carriers, their interaction with an external coherent electromagnetic field, and the renormalization effects associated with electron-hole interaction. Within the present approach, these features are taken fully into account.

When trying to generalize the physically intuitive picture of a semiclassical simulation consisting of random free flights, scattering events, and generation processes to the case of coherent phenomena, several difficulties occur. In the coherent regime the generation of carriers is a two-step process: First, the light field creates an interband polarization, then this polarization interacts again with the light field creating carriers. Thus, in addition to the Boltzmann equations, the equation of motion for the polarization has to be solved. Furthermore, the resulting generation rate is not positive definite, which means that recombination processes necessarily have to be taken into account. Within a "conventional" ensemble Monte Carlo calculation, where each carrier i is characterized by its wave vector $\mathbf{k}_i$, this is quite complicated. For each recombination process the states of all carriers have to be checked in order to find a pair with vanishing total momentum which can perform the recombination. This problem is related to the fact that we have specified more information than necessary. Therefore we describe the state of the carrier ensemble by means of a "number representation," obtained by introducing a phase-space discretization and specifying only the number of carriers in a cell. This description provides a clear and simple treatment of generation and recombination processes and is similar to recent cellular automata type calculations for semiclassical transport [18].

The basic lines of the simulation proceed as follows: The total time is divided into time steps. At the beginning of each time step, the system is completely specified by the distribution functions and polarizations. From these quantities we calculate the self-energy, the internal field, and the resulting generation rate. New electron-hole pairs are then created or removed accordingly. The polarization is obtained by numerical integration. The equations for the distribution functions are solved by means of a standard Monte Carlo technique [7]. Since the number of carriers in a cell is known at any time, the correct final-state occupation in the scattering rates can be used. Thus, the number of carriers never exceeds the limit given by the total number of states in the cell which is important for simulations close to degeneracy.

First, we will discuss some numerical results concerning simulations characterized by a laser energy far from the band gap. In Fig. 1 the generation rates obtained from the Monte Carlo simulation (integrated over the angular variables) are shown as a function of the wave vector k for different times during the laser pulse. Figure 1(a) refers to the full generation model while 1(b) refers to the semiclassical model. The latter ones, used in a conventional Monte Carlo simulation, are strictly positive. On the other hand, the rates in 1(a) exhibit a strong time dependence also in the shape. At short times the shape of the generation rate is much broader than estimated from the uncertainty principle. The narrowing with increasing time is accompanied by the buildup of negative regions off resonance which can be interpreted as a stimulated emission process. Thus, the distribution of the generated carriers becomes narrower not only due to a generation mainly in resonance but also due to a recombination of those carriers generated off resonance at short times.

In Fig. 2 the mean kinetic energies of electrons and holes [2(a)] and the polarizations [2(b)] are shown as functions of time. The energy of the holes is practically independent of the generation model while the electron energy, in particular during the pulse, is increased in the retarded model. This can be understood from the energy uncertainty in the generation rate. The broadening at short times is symmetric in energy with respect to the laser frequency. The density of states, however, increases with increasing energy, thus it is more probable to generate carriers above resonance than below. Of course, the stimulated emission then also preferably removes these carriers [see Fig. 1(a), dotted line]. Therefore, after the end of the pulse the difference is relatively small. However, the energy remains slightly larger during the whole simulation.

In contrast to the kinetic energies, the polarizations cannot be obtained from the solution of the Boltzmann equation, but necessarily require a solution of the coupled system of Bloch equations as performed in our simulations. In Fig. 2(b) the solid curve refers to the absolute value of the total polarization. It decays due to inhomogeneous broadening in k space, since each contribution rotates with a different frequency. The dashed curve refers to the incoherently summed polarizations. It decays due to incoherent scattering processes and the relaxation time is of the order of 0.5 ps. From an experimental point of view, the decay of the incoherently summed polarization is related to the photon-echo experiments [9].

Since the kinetic energies shown above are not directly accessible by experiments, in Fig. 3 the luminescence

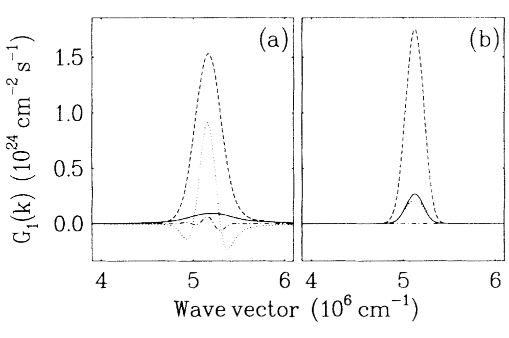

FIG. 1. Generation rate for a 0.1-ps pulse as a function of the wave vector k at different times: (a) with the full model and (b) in the classical limit. The curves refer to the times 0.1 ps (solid line), 0.2 ps (dashed line, the center of the pulse), 0.3 ps (dotted line), and 0.4 ps (dash-dotted line). Case (a) shows a narrowing and an associated recombination off resonance, while the classical rate is always positive.

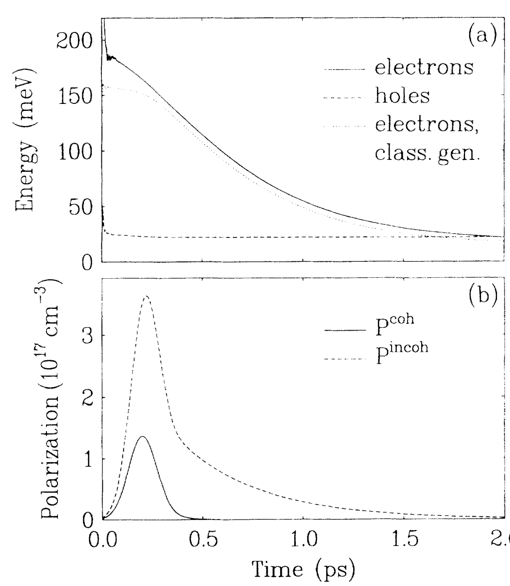

FIG. 2. (a) Kinetic energies of electrons and holes and (b) polarizations as functions of time. For the case of electrons the energy resulting from a semiclassical simulation is also shown. Because of the larger effective mass the energy of the holes does not exhibit significant differences.

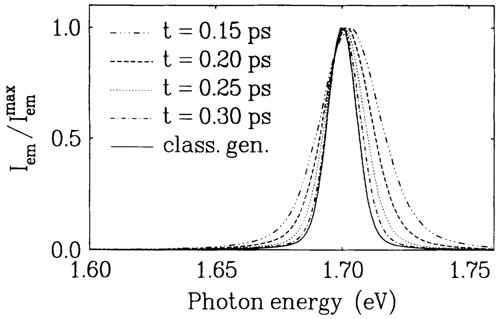

FIG. 3. Luminescence spectra at different times during the presence of the laser pulse (the center of the pulse is at 0.2 ps). The spectra are normalized to their maxima in order to emphasize the changes in their shape. Using the classical generation model, the shape does not depend on time and the resulting spectrum is given by the solid line. Within the full generation model, the spectra reflect the time dependence of the broadening of the generation rate [Fig. 1(a)].

spectrum as a function of the photon energy is shown. We have shown the very early stage of the process, when the laser is still active. Luminescence experiments are performed already at these times [4] where the models show remarkable differences. In a semiclassical treatment, the linewidth of the luminescence spectrum is independent of time (solid line). On the contrary, using the full generation model, we observe a strong time dependence of the luminescence line shape, which approaches the semiclassical result towards the end of the pulse. From this result, we see that, for the analysis of spectra at ultrashort times, coherent effects in the generation process should not be neglected.

Within the semiclassical generation rate, no strongly degenerate conditions can be reached and, at most, a half filling of the bands can be obtained. On the contrary, a coherent light field can produce a complete inversion. Therefore we have performed a simulation where the laser amplitude has been increased by a factor of 500. The kinetic energies and polarizations as functions of time are plotted in Fig. 4. The inset shows the carrier density during the pulse as a function of time. Now Rabi oscillations during the presence of the pulse are clearly visible. We want to remark that this simulation is not intended to represent a realistic situation (in particular due to the absence of carrier-carrier scattering processes), rather it demonstrates that the present approach works even under these extreme conditions.

Finally, we come to the analysis of the numerical results concerning laser excitations close to and within the band gap. This is the region where the effects due to interband Hartree-Fock terms, not accessible by a "conventional" Monte Carlo simulation, are most pronounced. Figure 5 shows the carrier density generated by a laser pulse with fixed amplitude and pulse width as a function of the excess energy. Both curves are obtained within the full generation model, for the solid curve the Hartree-

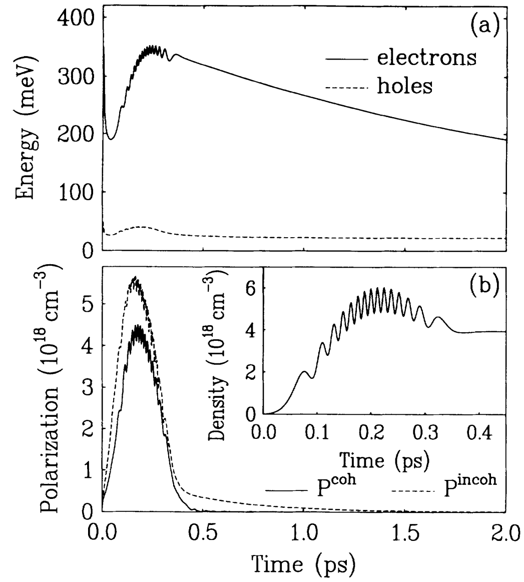

FIG. 4. (a) Kinetic energies of electrons and holes and (b) polarizations as functions of time for the strongly degenerate case. Inset: The density as a function of time during the pulse. The coherent light-matter interaction leads to Rabi oscillations in the density and associated oscillations in the electronic energy and polarizations.

Fock terms have been included while the dashed curve refers to a simulation without these terms. As expected, without taking into account the carrier-carrier interaction, the density is practically zero for excitation below the gap. Including Hartree-Fock terms, we recover the strong $n=1$ exciton line and a subsequent minimum between the $n=1$ and $n=2$ exciton lines. These results confirm the treatment of Hartree-Fock terms within our numerical simulation.

In conclusion, we have presented a numerical method which enabled us to include coherent phenomena in a

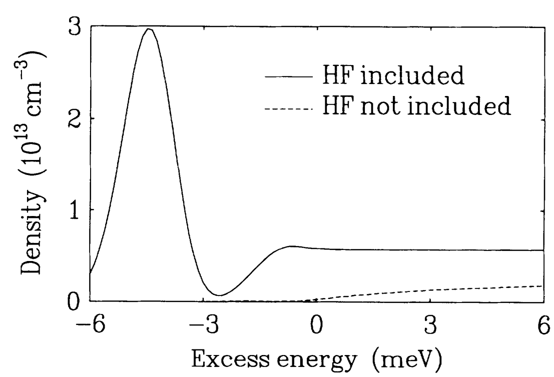

FIG. 5. Carrier density created by a laser pulse with fixed amplitude and a pulse width of 1 ps as a function of the excess energy of the laser. Without taking into account the carrier-carrier interaction, the density is practically zero for excitation below the gap. With Hartree-Fock terms excitonic effects appear.

Monte Carlo simulation. The present approach allowed us to include phenomena not accessible by a "conventional" Monte Carlo analysis: (i) The explicit treatment of the polarization gives information on the dephasing of the system. (ii) The generation process is treated in a self-consistent way with its full time-dependent line shape. (iii) Because of the Hartree-Fock terms the region close to the band gap also can be studied, where excitonic features cannot be neglected.

For a quantitative analysis of experimental results carrier-carrier scattering processes will have to be included. Within the present approach this does not constitute a principle problem. The results shown here, however, have demonstrated that coherent phenomena in the theoretical analysis of ultrafast processes in semiconductors should not be neglected, even if only "incoherent" properties like carrier thermalization are studied.

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