Ultrafast Relaxation of Photoexcited Carriers: The Role of Coherence in the Generation Process

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A self-consistent description of the ultrafast dynamics of photoexcited carriers in semiconductors based on a generalized Monte Carlo solution of the semiconductor Bloch equations is presented. The problem of photogeneration and its theoretical description are discussed. We show that some of the approaches commonly used fail in describing correctly the effect of carrier-carrier interaction in the low-density limit. By including terms which have the structure of "in-scattering" terms (vertex corrections) for the interband polarization, the experimentally observed features in the carrier dynamics are well described in the whole density range.

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The study of the ultrafast dynamics of optically excited carriers in semiconductors provides valuable information on the role of the various interaction mechanisms for the thermalization and relaxation process [1–10]. By varying the carrier density, the relative weight of the different scattering mechanisms also changes: At high densities carrier-carrier scattering leads to an extremely fast broadening of the distribution function and no particular structures are observed [2–4]; at low densities the interaction with optical phonons is the dominant mechanism leading to the formation of phonon replicas in the carrier distribution function [5,6]. In the latter case, for excitation below the threshold for optical phonon emission very long thermalization times have been observed [7].

The theoretical analysis of these experiments is usually based on the semiclassical Boltzmann transport equations (BTE) for the distribution functions of electrons and holes. They are solved by means of Monte Carlo (MC) simulations [2–4,10–13] which turned out to be the best suited technique to include on a kinetic level a large variety of different scattering processes. The main reason for the experimentally observed strong density dependence is carrier-carrier scattering. Therefore, much work has been devoted to the theoretical modeling of the different aspects of this complicated many-body problem involving exchange effects [14], plasmon-phonon coupling [15,16], and the static or dynamic treatment of screening [3,4,17–20].

On the semiclassical level the coherence of the light field and the resulting coherent polarization in the carrier system is completely neglected. As shown in many experiments, this coherence can be directly measured, e.g., by means of four-wave-mixing experiments [21–24] or by studying the coherent emission of submillimeter radiation [25,26]. However, as recently pointed out in several theoretical works [17,27–29], the progressive reduction of the typical time scale leads to physical conditions where such coherent phenomena become important also in energy-relaxation experiments. In such conditions, a proper theoretical approach has to include both classes of phenomena on the same kinetic level.

One of the crucial points in the description of a relaxation experiment in the subpicosecond regime is the treatment of the carrier photogeneration process. In the semiclassical kinetic picture, which is the basis of conventional MC simulations [3,30], the carrier dynamics is governed by the BTE for the distribution functions of electrons and holes. The interaction with the light field is treated in the same way used for other scattering processes: The carriers are generated according to an electron-hole generation rate obtained from Fermi's golden rule. The energy broadening is usually introduced as a phenomenological parameter which is estimated from the pulse duration on the basis of the energy-time uncertainty relation. Therefore, such a description of the generation process does not provide any information about the time dependence of its spectral width.

On the contrary, a self-consistent description of the generation process can be obtained within the framework of the semiconductor Bloch equations (SBE) [27,29,31,32]. Here, in addition to the distribution functions of electrons and holes, the interband polarization $p_{\mathbf{k}}$ is considered as an independent variable. In this formulation, the generation process is a two-step process: First, the light field creates a polarization, and second, carriers are generated due to the coupling between the light field and the polarization. This approach leads directly to the correct spectral and temporal dependence of the generation rate [29].

However, within this more rigorous approach the en-

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ergy broadening of the generated carrier distribution strongly depends on the model employed for the description of the electron-hole dephasing process. This dephasing due to incoherent scattering processes is usually described in terms of a dephasing rate $\Gamma_{\mathbf{k}}$, the imaginary part of the self-energy [17,29], according to

$$\frac{d}{dt}p_{\mathbf{k}}\bigg|_{\text{inco}} = -\Gamma_{\mathbf{k}}p_{\mathbf{k}}. \tag{1}$$

This k-dependent dephasing rate is directly related to the scattering rates of electrons and holes [17,29]. In order to simplify the calculations, the $\mathbf{k}$ dependence is often neglected resulting in a dephasing time $T_2 = \Gamma^{-1}$ [24,33]. As is well known and will be discussed in detail below, for the case of carrier-carrier scattering this model leads to a completely wrong density dependence of the relaxation process. We never find an appropriate decreasing of the energy broadening of the generated carriers with decreasing density.

However, in the microscopic derivation of the SBE additional incoherent contributions to the polarization dynamics appear [27,34,35]. The aim of this paper is to show that the failure in describing the correct density dependence is simply due to the neglect of these additional contributions. What we propose is the use of a more refined model of electron-hole dephasing which results in a kinetic equation for the interband polarization describing the incoherent scattering processes on the same microscopic level as for the usual BTE. The model is obtained within the following approximation scheme: (i) We limit ourselves to contributions up to second order in the interaction matrix elements, (ii) the second-order terms are treated within the usual Markov approximation neglecting the real parts which give corrections to the Hartree-Fock self-energies [29], and (iii) all second-order contributions which involve second or higher powers of the polarization ("polarization scattering") are neglected [35,36]. These are approximations generally made in the derivation of the incoherent part of the SBE where, in particular due to (iii), all excitonic effects in the scattering rates are neglected. Since here we are mainly interested in energy regions far from the band gap, such effects can be assumed to play a minor role. Within these approximations, the various incoherent contributions entering the equation of motion for the interband polarization result to have the structure of rate equations of the general form [27]

$$\frac{d}{dt}p_{\mathbf{k}}\bigg|_{\text{inco}} = \sum_{\mathbf{k}'}\left[s_{\mathbf{k},\mathbf{k}'}p_{\mathbf{k}'} - s_{\mathbf{k}',\mathbf{k}}p_{\mathbf{k}}\right] . \tag{2}$$

Compared with Eq. (1), it contains in addition what in the Boltzmann theory are called the "in-scattering terms" (vertex corrections in the language of Green's-function theory). They are usually neglected on the basis of a random phase argument: Since the polarization is a complex quantity, it is assumed that different $\mathbf{k}'$ contributions in the first term cancel leading to Eq. (1) with $\Gamma_{\mathbf{k}} = \sum_{\mathbf{k}'} s_{\mathbf{k}',\mathbf{k}}$. We will show that this assumption is not fulfilled if, as for the case of low densities, small-angle scattering is strongly dominant.

Within the present approach, the incoherent contribution to the polarization dynamics has exactly the same formal structure as the "Boltzmann collision term" in the equations for the distribution functions. On the basis of this strong similarity, a further extension of the generalized Monte Carlo method described in [28,29] has been developed. It provides a solution of our SBE by means of a full MC sampling of the above incoherent contributions for distribution functions as well as for the polarization. The main difference between the polarization and the distribution functions lies in the fact that the former is a complex quantity, while the latter are positive-definite quantities. However, this does not limit the use of MC simulations as already pointed out in Ref. [24], where the complex Fourier components of the distribution functions in a four-wave-mixing experiment have been calculated by means of MC simulations.

In order to address the physical problem discussed above, we have performed several "simulated experiments." For all these simulations, a two-band semiconductor model has been considered: The carrier-phonon interaction has been introduced in terms of both acoustic and polar optical phonon scattering while carrier-carrier interaction has been treated in terms of a time-dependent statically screened Coulomb potential. The carriers are generated by a laser pulse with Gaussian temporal shape (pulse duration $\tau_L = 50$ fs, excess energy $\mathcal{E}_{\text{ex}} = 0.18\,\text{eV}$, pulse maximum at $t = 0$). The three different approaches discussed above have been investigated: (a) the Boltzmann case where the generation rate exhibits a Gaussian energy broadening according to the Gaussian broadening in time, (b) the Bloch case with only dephasing processes according to Eq. (1), and (c) the Bloch case including in-scattering terms for the polarization according to Eq. (2).

In Fig. 1 the electron distribution at $t = 100$ fs is plotted as a function of energy for different densities. In the Boltzmann case [Fig. 1(a)] the broadening of the generation rate is independent of the density. At the lowest density we clearly see the phonon replicas of the initial peak. With increasing density, the increasing efficiency of carrier-carrier scattering leads to an increase in the broadening and at the highest density there are no more structures related to the phonon emission. However, we still find a quite pronounced decay of the distribution function at high energies. In the Bloch case, by considering only dephasing processes according to Eq. (1), the situation changes completely [Fig. 1(b)]. The distribution function is found to be very broad and nearly independent of the density. No phonon replicas are visible. This is in clear contradiction to experimental results where phonon replicas are found in the low-density limit. Here, the broadening in the generation process is governed by dephasing rate $\Gamma_{\mathbf{k}}$ since it is larger than the inverse of the pulse duration. It is nearly density independent due to

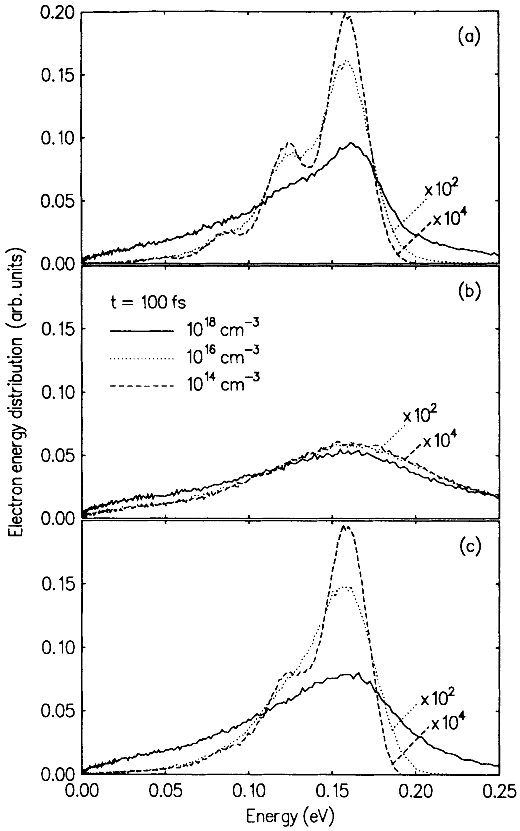

FIG. 1. Electron distribution as a function of energy at $t = 100$ fs for the case of generation with a 50 fs laser pulse for different final carrier concentrations, obtained (a) from the Boltzmann equations, (b) from the Bloch equations without in-scattering terms, and (c) from the Bloch equations including in-scattering terms.

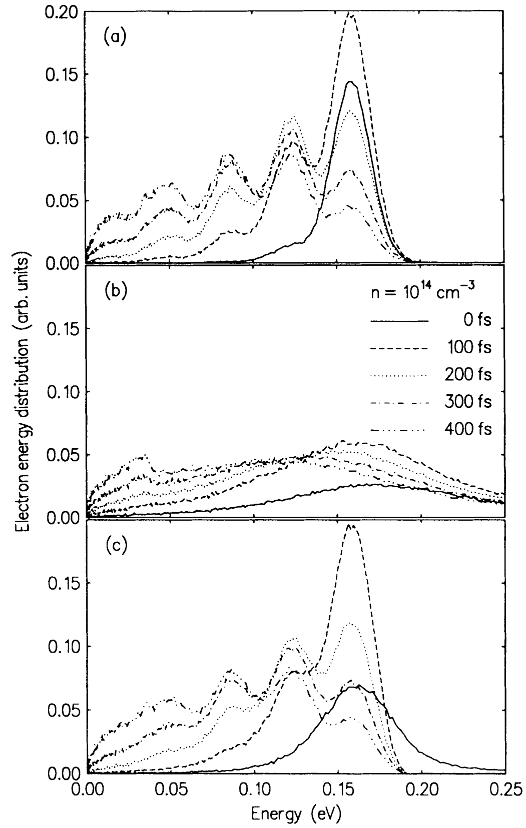

FIG. 2. Electron distribution as a function of energy at different times for the case of generation with a 50 fs laser pulse in the low-density limit, obtained (a) from the Boltzmann equations, (b) from the Bloch equations without in-scattering terms, and (c) from the Bloch equations with in-scattering terms.

the cancellation of two effects: With decreasing density the number of scattering partners for carrier-carrier scattering decreases; however, due to the decreasing screening wave vector, the interaction extends over a larger volume. Therefore, the total scattering rate remains approximately constant, but the scattering angle and the energy exchange in a scattering process decrease. This latter effect does not influence $\Gamma_{\mathbf{k}}$; however, it strongly influences the in-scattering contribution in Eq. (2) as shown in Fig. 1(c). The broadening in the low-density limit is strongly reduced and we recover the phonon replicas as observed in the Boltzmann case. In the high-density limit it turns out that the distribution function is nearly unchanged with respect to case (b). This justifies the use of the total scattering rate as a dephasing rate. The reason is that in this case, due to the strong screening, the energy transfer in a scattering process is large. Thus, the in-scattering term connects polarization components with strongly different wave vectors and the random-phase assumption is well justified. As observed experimentally, the distribution is much broader than obtained from the Boltzmann equation and, in particular at high energies, it does not exhibit the pronounced decay of case (a). While in a semiclassical model the carriers are generated in a relatively narrow energy range and subsequently the distribution is broadened due to scattering processes, here already the generation rate is strongly broadened by the dephasing mechanisms.

Figure 2 shows the electron distribution at different times in the low-density limit (final carrier density $n = 10^{14}$ cm^{-3}). In the Boltzmann case we see the buildup of the phonon replicas. With increasing time, they become slightly broadened due to carrier-carrier scattering which, however, is not very efficient at this low density. Again, in the Bloch case without in-scattering contributions the distributions are extremely broad exhibiting no phonon-related structures, which is clearly unphysical at these low densities. Taking into account the in-scattering terms we find the initial broadening during the pulse related to the energy-time uncertainty. Then, we recover the buildup of the phonon replicas with an increased broadening compared to the Boltzmann case which is reminiscent of the dephasing process during the carrier generation. In the derivation of the semiclassical generation rate this dephasing is neglected since the rate for each interaction mechanism is calculated independently.

In conclusion, we have discussed the strong limitations of the conventional theoretical approaches used in describing the ultrafast dynamics of photogenerated carriers in subpicosecond energy-relaxation experiments. In particular, we have shown the inadequacy of the phenomenological electron-hole dephasing models in describing the role of carrier-carrier interaction in the low-density limit. We have then demonstrated how this pathological situation can be completely removed using a formulation of the problem in which the incoherent scattering processes are treated at the same microscopic level both for the distribution functions and for the interband polarization. In this case, the results exhibit the typical features of the carrier dynamics at low densities, i.e., the formation of phonon replicas, as well as at high densities, i.e., a large initial energy broadening.

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