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A microscopic quantum-kinetic theory based on density matrix formalism is formulated to describe the processes of short pulse laser interaction with materials such as semiconductors accounting for arbitrary spatial inhomogeneities in the excitation conditions and other spatial phenomena such as filamentation of tightly focused femtosecond laser pulses, structural modification and catastrophic optical damage. A system of Boltzmann-Bloch transport equations are established that include both space and momentum dependence of the electron and hole distribution functions and the polarization. Microscopic electronphonon and electron-electron scattering terms as well as scattering terms that lead to transitions between valence and conduction bands, i.e. impact ionization and recombination terms, are included explicitly in the equations. The formulated theory describes the spatio-temporal dynamics of electrons and holes in inhomogeneously excited materials including the coherent interactions of carriers and the laser light field as well as transport due to spatial gradients and electrostatic forces.

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Report on proposal “Theoretical modeling of ultrashort laser pulse interaction with dielectric and semiconductor materials”

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A quantum-kinetic theory has been formulated to describe the spatiotemporal microscopic carrier dynamics in laser excited semiconductor materials that accounts for the effects of inhomogeneous excitation and structural inhomogeneities due to bulk fillamentation damage and micro/nano structuring by introducing space dependent carrier distributions and coherency. Two alternative approaches to the problem have been presented - one is a non-trivial generalization of our previously derived Fokker-Planck equation in energy space, and the other is density matrix based formalism. Both approaches involve extensive computational effort.

I. NON-LOCAL FOKKER-PLANCK EQUATION FOR LASER-INDUCED BREAKDOWN IN MATERIALS

A. Introduction

Ultra-short laser-induced breakdown studies have revealed the possibility of self-channeling ultrafast light in condensed media with the strong involvement of single or multiphoton ionization and avalanche ionization capable of delivering high free-electron densities. The full description of the laser-matter interaction process leading to material modification, and the complex spatio-temporal dynamics of the beam propagation in the media from first principles comprises the nonlinear Schrödinger equation for the laser electric field envelope coupled with an equation describing the growth of the free-electron density $n(\vec{r},t)$ [1], [2]. Usually the equation for $n(\vec{r},t)$ employed in the literature is a simple rate equation not accounting for the energy absorption process from the laser field. The slowly varying [1] envelope function $\vec{\mathrm{E}}$ of the electric field of the laser $\vec{E}$ is defined by: $\vec{E}\left(\vec{r},t\right)=\sqrt{\frac{\Omega_L\mu_0}{2k_0}}\vec{\mathrm{E}}\left(\vec{r},t\right)e^{i(k_0z-\Omega_0t)}+c.c.$ (for a pulsed beam centered on the operating frequency Ω_L, wave number k_0); we choose the electromagnetic radiation to be polarized in the x direction $\vec{\mathrm{E}}=(\mathrm{E},0,0)$. The free electrons motion is governed by the oscillation of the inhomogeneous optical field beating at Ω_L so that an electron current density with a slowly varying envelope induces a variation of the local population of electrons in space and time leading to a space-dependent carrier distribution. Since the time development of the avalanche ionization process depends also very strongly on the values of the field, it follows that at any spatially non-uniform distribution of the electromagnetic radiation density, initial electron density gradients ∇_r could occur that may give rise to more or less strong diffusion currents. Therefore, instead of using a simple rate equation for the electron density $n(\vec{r},t)$ produced by the laser field, we augment phenomenologically the kinetic equation previously derived by us for the electron energy distribution function to account for its spatial dependence when the laser field is spatially inhomogeneous. The rates in the equation explicitly include the effect of laser field energy absorption by the free electrons.

B. Theoretical model

A kinetic equation is derived for the conduction-electron distribution function following references [3, 4, 5]. We assume that the frequency of the electromagnetic field satisfies the conditions $1 \ll \Omega_L \tau_p \ll c/\bar{v}$ (the electromagnetic field frequency exceeds appreciably the frequency of electron-phonon collisions), $\Omega \gg \omega_p$, $\bar{\varepsilon}\tau_p \gg 1$ (where τ_p is the momentum relaxation time, ω_p is the electron plasma frequency and $\bar{\varepsilon}$ and $\bar{v}$ are the average energy and velocity of the electron). These conditions are sufficient to guarantee the penetration of the electromagnetic field deeply into the semiconductor and also mean that the wavelength is much larger than the mean-free-path or the De Broglie wavelength of an electron, the wavelength of the phonons which interact with the electrons and allow for the use of dipole approximation of the radiation field. They imply that the dependences of these processes on the electromagnetic field are local. Under the same conditions, the magnetic field contribution to the interaction with the laser field is negligible because it is of the same order of magnitude as the electric quadrupole contribution. We first consider an electron-phonon system in a spatially uniform sinusoidal electric field with a vector potential $A(t) = \mathrm{A}\sin(\Omega_L t)$ and find the solution of the Schrödinger equation

$$i\hbar \frac{\partial \psi(\vec{r},t)}{\partial t} = \frac{1}{2m^*}\left[\vec{p} + \frac{e}{c}\vec{A}\left(\vec{r},t\right)\right]^2 \psi\left(\vec{r},t\right). \tag{1}$$

Using the standard time-dependent perturbation theory and the calculated electron wave function renormalized by the laser field, we calculate the electron transition rate. Unlike the isotropic transition rate obtained in the absence of the laser field, the presence of linearly polarized electromagnetic radiation results in an anisotropic electronic transition rate characterized by field dependence via a Bessel function term – this effect is known as free-electron absorption of photons. For moderately high intensities of the laser radiation, the Bessel function is expanded to second order. In the diffusive limit $\hbar\omega_q \ll E_k^e$, where $E_k^e = \hbar^2 k^2 / 2m*_e$ and $\hbar\omega_q$ are the free electron kinetic energy in the conduction band with effective mass m_e^* and the phonon energy respectively, the electron distribution function is expanded in Taylor series. Thus the linearized Fokker-Plank type equation is obtained. The classical Joule heating of electrons from the field which is conspicuously missing in other quantum-kinetic approaches is included by taking into account the local fluctuation of the electron kinetic energies. Source terms such as stimulated interband electron transitions due to a single or multiphoton absorption, impact ionization due to Coulomb interaction between electrons and holes and sink terms such as non-radiative recombination due to a phonon-mediated interaction are included in the equation:

$$\frac{\partial f^e(E,t)}{\partial t} + V(E,t)\frac{\partial f^e(E,t)}{\partial E} - D(E,t)\frac{\partial^2 f^e(E,t)}{\partial \left(E_k^e\right)^2} = A(E,t)\, f^e(E,t) + S(E,t). \tag{2}$$

The rates in equation (2) are given by the following expressions [3], [4]:

$$V\left(E,t\right)=V_T\left(E\right)+V_F\left(E\right)+\frac{1}{3}\sigma\left(\Omega_L\right)\mathrm{E}^2+A_T(E)\frac{\tau_p}{3}\sigma\left(\Omega_L\right)\mathrm{E}^2, \tag{3}$$

$$D\left(E,t\right)=D_T\left(E\right)+D_F\left(E\right)+\frac{2}{3}\sigma\left(\Omega_L\right)\mathrm{E}^2E_k^e+V_T(E)\frac{\tau_p}{3}\sigma\left(\Omega_L\right)\mathrm{E}^2, \tag{4}$$

where

$$V_T\left(E_{\vec{k}}\right)=\frac{2\pi}{\hbar}\sum_{\vec{q}\,\lambda}\left|C_{\vec{q}\,\lambda}\right|^2\hbar\omega_{\vec{q}\,\lambda}\times\left[N_{\vec{q}\,\lambda}^{ph}\delta\left(E_{\vec{k}}-E_{\vec{k}+\vec{q}}-\hbar\omega_{\vec{q}\,\lambda}\right)-\left(N_{\vec{q}\,\lambda}^{ph}+1\right)\delta\left(E_{\vec{k}}-E_{\vec{k}+\vec{q}}+\hbar\omega_{\vec{q}\,\lambda}\right)\right], \tag{5}$$

$$\begin{aligned}V_F=&\frac{2\pi}{\hbar}\sum_{\vec{q}\,\lambda}\left|C_{\vec{q}\,\lambda}\right|^2\Im_{\vec{q}}^2\times N_{\vec{q}\,\lambda}^{ph}(E_k-E_{k-q})\left[\delta\left(E_{\vec{k}}-E_{\vec{k}-\vec{q}}-\hbar\omega_{\vec{q}\,\lambda}+\hbar\Omega_L\right)+\delta\left(E_{\vec{k}}-E_{\vec{k}-\vec{q}}-\hbar\omega_{\vec{q}\,\lambda}-\hbar\Omega_L\right)\right]\\&+\left(N_{\vec{q}\,\lambda}^{ph}+1\right)(E_k-E_{k-q})\left[\delta\left(E_{\vec{k}}-E_{\vec{k}+\vec{q}}+\hbar\omega_{\vec{q}\,\lambda}+\hbar\Omega_L\right)+\delta\left(E_{\vec{k}}-E_{\vec{k}+\vec{q}}+\hbar\omega_{\vec{q}\,\lambda}-\hbar\Omega_L\right)\right],\end{aligned} \tag{6}$$

$$D_T(E_{\vec{k}})=\frac{\pi}{\hbar}\sum_{\vec{q}\,\lambda}|C_{\vec{q}\,\lambda}|^2(\hbar\omega_{\vec{q}\,\lambda})^2\times\left[N_{\vec{q}\,\lambda}^{ph}\delta(E_{\vec{k}}-E_{\vec{k}+\vec{q}}-\hbar\omega_{\vec{q}\,\lambda})-(N_{\vec{q}\,\lambda}^{ph}+1)\delta(E_{\vec{k}}-E_{\vec{k}+\vec{q}}+\hbar\omega_{\vec{q}\,\lambda})\right], \tag{7}$$

$$\begin{aligned}D_F=&\frac{\pi}{4\hbar}\sum_{\vec{q}\,\lambda}|C_{\vec{q}\,\lambda}|^2\Im_{\vec{q}}^2\times N_{\vec{q}\,\lambda}^{ph}(E_k-E_{k-q})^2\left[\delta(E_{\vec{k}}-E_{\vec{k}-\vec{q}}-\hbar\omega_{\vec{q}\,\lambda}+\hbar\Omega_L)+\delta(E_{\vec{k}}-E_{\vec{k}-\vec{q}}-\hbar\omega_{\vec{q}\,\lambda}-\hbar\Omega_L)\right]\\&+(N_{\vec{q}\,\lambda}^{ph}+1)(E_k-E_{k-q})^2\left[\delta(E_{\vec{k}}-E_{\vec{k}+\vec{q}}+\hbar\omega_{\vec{q}\,\lambda}+\hbar\Omega_L)+\delta(E_{\vec{k}}-E_{\vec{k}+\vec{q}}+\hbar\omega_{\vec{q}\,\lambda}-\hbar\Omega_L)\right].\end{aligned} \tag{8}$$

The right-hand side of (1) contains the coefficients for thermal spontaneous phonon emission and the one for the field-induced phonon emission.

$$A_T(E_{\vec{k}})=\frac{2\pi}{\hbar}\sum_{\vec{q}\,\lambda}|C_{\vec{q}\,\lambda}|^2\times\left[N_{\vec{q}\,\lambda}^{ph}\delta(E_{\vec{k}}-E_{\vec{k}+\vec{q}}-\hbar\omega_{\vec{q}\,\lambda})-(N_{\vec{q}\,\lambda}^{ph}+1)\delta(E_{\vec{k}}-E_{\vec{k}+\vec{q}}+\hbar\omega_{\vec{q}\,\lambda})\right], \tag{9}$$

$$\begin{aligned}A_F=&\frac{\pi}{2\hbar}\sum_{\vec{q}\,\lambda}|C_{\vec{q}\,\lambda}|^2\left(e|\vec{q}.\vec{\mathrm{E}}(t)|/m^*\Omega_L^2\right)^2\times\left[\delta(E_{\vec{k}}-E_{\vec{k}-\vec{q}}-\hbar\omega_{\vec{q}\,\lambda}+\hbar\Omega_L)+\delta(E_{\vec{k}}-E_{\vec{k}-\vec{q}}-\hbar\omega_{\vec{q}\,\lambda}-\hbar\Omega_L)\right]\\&+\left[\delta(E_{\vec{k}}-E_{\vec{k}+\vec{q}}+\hbar\omega_{\vec{q}\,\lambda}+\hbar\Omega_L)+\delta(E_{\vec{k}}-E_{\vec{k}+\vec{q}}+\hbar\omega_{\vec{q}\,\lambda}-\hbar\Omega_L)\right].\end{aligned} \tag{10}$$

In these expressions E denotes the amplitude of the laser field, the indices T and F denote the thermal contribution due to phonon scattering and field induced contribution respectively, $\left|C_{\vec{q}\,\lambda}\right|$ is the electron-phonon coupling and $\Im_{\vec{q}}=\left(e\left|\vec{q}.\vec{\mathrm{E}}\left(t\right)\right|\Big/m^*\Omega_L^2\right)$. The expressions for the field dependent rates A_F, V_F and D_F reveal that in the presence of electromagnetic radiation the electron-phonon scattering can be accompanied by electronic transitions through photon absorption and emission.

C. Generalized Fokker-Planck kinetic equation

By explicitly substituting the spatially dependent envelope of the laser $\vec{\mathrm{E}}\left(\vec{r},t\right)$ field in equations (3)–(10) the space and energy dependent rates $V\left(\vec{r},E,t\right)$ and $D\left(\vec{r},E,t\right)$ are obtained. Equation (2) is different from the standard Fokker-Planck equation since the derivation does not make it a conservation equation of the electron number and current density even in the absence of the source terms. We define the current in energy space:

$$J^{E}\left(E,t\right)=V\left(E,t\right)f^{e}\left(E,t\right)-\frac{\partial\left[D(E,t)f^{e}\left(E,t\right)\right]}{\partial E},$$

and equation (2) is written as

$$\frac{\partial f^{e}\left(E,t\right)}{\partial t}+\frac{\partial J^{E}\left(E,t\right)}{\partial E}+2\frac{\partial D\left(E,t\right)}{\partial E}\frac{\partial f^{e}\left(E,t\right)}{\partial E}=A\left(E,t\right)f^{e}\left(E,t\right)+\tilde{A}\left(E,t\right)f^{e}\left(E,t\right)+S(E,t),\quad(11)$$

where $\tilde{A}(E,t)=\dfrac{\partial V(E,t)}{\partial E}-\dfrac{\partial^{2}D(E,t)}{\partial E^{2}}$.

For the time being we ignore the third term on the left-hand side of equation (11). Bearing in mind that the Joule heating $V\left(\vec{r},E,t\right)$ and diffusion $D\left(\vec{r},E,t\right)$ rates of equation (2) already include space dependence, a generalized current $J^{0}\left(\vec{r},E,t\right)$ is defined reflecting the energy gain or loss by electrons. The electron distribution function is also generalized to include space dependence

$$J^{0}\left(E,\vec{r},t\right)=V\left(E,\vec{r},t\right)f\left(E,\vec{r},t\right)-\frac{\partial\left[D(E,\vec{r},t)f\left(E,\vec{r},t\right)\right]}{\partial E}.$$

In references [6] and [7], a theory of electron drift and diffusion was proposed allowing for a non-equilibrium energy distribution, such as occurs in high electric fields. The transport equation assumes a form of a Fokker-Planck equation in a four-dimensional (4D) energy-position continuum instead of the 3D position space. In this equation the 4D current density is a sum of a drift and a diffusion terms that depend on the $(E,\vec{r})$ point. The use of this equation assumes that not only the energy but also the position coordinates vary continuously, therefore demanding that the electron energy be exchanged with the lattice in small steps (e.g. phonons of energy $\hbar\omega_{q}\ll E$) and that electron velocity be finite.

Following [6], electron transport in an inhomogeneous laser field involving continuous energy and coordinate changes is described by an equation of continuity in the energy-position manifold. So we generalize equation (11) as

$$\frac{\partial f^{e}\left(E,\vec{r},t\right)}{\partial t}+\frac{\partial J^{0}\left(E,\vec{r},t\right)}{\partial E}+\frac{\partial J^{\beta}\left(E,\vec{r},t\right)}{\partial x^{\beta}}=\left[A\left(E,\vec{r},t\right)+\tilde{A}\left(E,\vec{r},t\right)\right]f^{e}\left(E,\vec{r},t\right)+S\left(E,\vec{r},t\right)\quad(12)$$

by adding currents in real space, where the Greek indices run from 1 to 3.

Due to our choice of laser field polarization in the x direction, we get for the currents in space:

$$J^x(E,\vec{r},t) = v_d(E,\vec{r},t)f^e(E,\vec{r},t) - \frac{\partial[e\mathrm{E}(\vec{r},t)D_x(E,\vec{r},t)f^e(E,\vec{r},t)]}{\partial E} - \frac{\partial[D_x(E,\vec{r},t)f^e(E,\vec{r},t)]}{\partial x}, \quad (13)$$

$$J^y(E,\vec{r},t) = -\frac{\partial[e\mathrm{E}(\vec{r},t)D_x(E,\vec{r},t)f^e(E,\vec{r},t)]}{\partial E} - \frac{\partial[D_x(E,\vec{r},t)f^e(E,\vec{r},t)]}{\partial y}, \quad (14)$$

$$J^z(E,\vec{r},t) = -\frac{\partial[e\mathrm{E}(\vec{r},t)D_x(E,\vec{r},t)f^e(E,\vec{r},t)]}{\partial E} - \frac{\partial[D_x(E,\vec{r},t)f^e(E,\vec{r},t)]}{\partial z}. \quad (15)$$

In equations (13)–(15), $\mathrm{E}(\vec{r},t)$ is the laser field envelope. We also generalize the expression for single photon excitation across the bandgap. The other two terms included in $S(E,\vec{r},t)$ – the impact ionization and the Auger recombination – are not affected directly by the laser field in our derivation.

$$S_{abs} \propto \frac{2\pi}{\hbar}|F_k|^2 \left[\frac{2F_k/\pi}{\left(\hbar\Omega_L - E_k^e - E_k^h - E_G\right)^2 + 4|F_k|^2}\right], \quad (16)$$

$$|F_k|^2 \approx \frac{e^2\mathrm{E}_{0L}^2(x,y,z,t)}{m_0\Omega_L}\left[\left(\frac{m_0}{m_e^*} - 1\right)\frac{E_G(E_G+\Delta_0)}{2(E_G+2\Delta_0/3)}\right]. \quad (17)$$

D. Relation between the rates in energy and coordinate space

Following closely references [8, 9, 10], we establish the connection between the real space drift velocity and the field dependent velocity in energy space which we have already derived. In this way we circumvent the necessity to introduce an expression for the energy dependent electron mobility in the given material and also use a quantum mechanically derived rate. We consider an electron with momentum $\vec{p}_0$ and energy $E(\vec{p}_0)$ at time $t = 0$. We define $\partial_F = e\vec{\mathrm{E}}\cdot\nabla_p$ where $\vec{\mathrm{E}}$ denotes the negative laser electric field. Then the energy change after $t = 0$ before any collision occurs is: $E(t) - E_0 = (\partial_F E)_{p_0} t + 1/2\left(\partial_F^2 E\right)_{p_0} t^2 + \ldots$ The probability for the electron starting from $\vec{p}_0$ not to experience collisions between 0 and t is $c(t) = \exp\left\{-\int_0^t dt'/[\tau(\vec{p})]\right\}$, where account is taken of the change in τ as the electron momentum changes in time as $d\vec{p}/dt = e\vec{E}$. Upon first order expansion of $\tau\left(\vec{p}_0 + e\vec{E}t\right)$, we obtain $c(t) = \exp(-t/\tau_0)\left[1 + 1/2(t/\tau_0)^2(\partial_F\tau)_{p_0}\right] + O\left(E^2\right)$, where $\tau_0 = \tau(\vec{p}_0)$.

The exponential gives the non-scattering probability if τ is constant while the electron is accelerated by the field and the second factor is the lowest-order correction due to the change in τ. We obtain the averages over a collision-free flight

$$\langle t\rangle = \tau_0\left[1 + (\partial_F\tau)_{p_0}\right] \text{ and } \left\langle t^2\right\rangle = 2\tau_0^2\left[1 + 3(\partial_F\tau)_{p_0}\right].$$

Then the average energy gained over a collisionless flight (of order E^2) is:

$$\langle E(t) - E_0\rangle = \left\{(\partial_F E)_{p_0} + \partial_F\left(\tau\partial_F^E\right)_{p_0}\right\}\tau_0 \text{ so } \langle E(t) - E_0\rangle = e\vec{E}\cdot\left\{\vec{v}_g(\vec{p}_0) + \left[\partial_F(\vec{v}_g\tau)_{\vec{p}_0}\right]\right\}\tau_0,$$

$$V_F(E_0) = \left(\tfrac{dE}{dt}\right)_F = e\vec{E}\cdot\left\{\vec{v}_g(\vec{p}_0) + \left[\partial_F(\vec{v}_g\tau)_{\vec{p}_0}\right]\right\}, \text{ with } \vec{v}_d(E_0) = \frac{\int\int_{E_0}\partial_F\left(\vec{v}_g\tau\left(\vec{p}\right)\right)dS_p/|\vec{v}_g|}{\int\int_{E_0}dS_p/|\vec{v}_g|},$$

where $dS_p/|\vec{v}_g|$ are the number of states in a surface element dS_p in momentum space. We thus finally obtain the relation between the field dependent drift velocity and the drift velocity in coordinate space: $V_F(E_0) = e\vec{E} \cdot \vec{v}_d(E_0)$ and $v_{d_x}(E, \vec{r}, t) = \frac{V_F(E,\vec{r},t)}{(e\mathrm{E}_{0L})}$. The position-space diffusion coefficient is related to the field dependent diffusion coefficient in energy space by [8]

$$D_x(E, \vec{r}, t) = \frac{D_F(E, \vec{r}, t)}{(e\mathrm{E})^2}.$$

Joule heating is the main energy gain mechanism, so the electrons from low-energy states in the conduction band are transferred to high-energy states by absorbing energy from the laser field. This is illustrated in figure 1 where the peak of the electron energy distribution function exhibits increased amplitude and is shifted to the right (higher electron energy) in the presence of the Joule heating term. Figure 2 shows the dependence of the electron average energy on the Joule heating term. It should be noted that the Joule heating term is only one contribution to the rate $V(E, t)$ (eq. 3) which has the physical meaning of energy dependent drift.

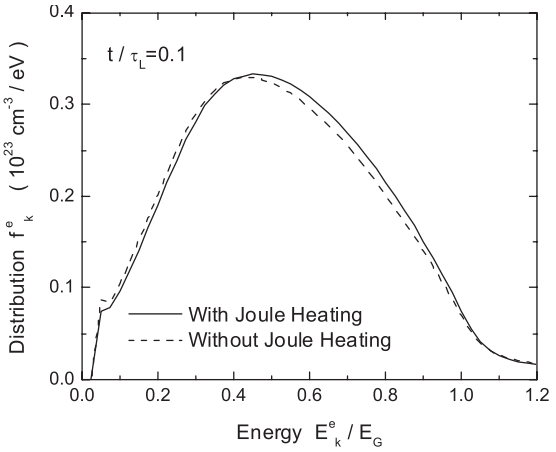

Figure 1. Comparison of the energy spectra of f_k^e at $t/\tau_L = -0.5$ including and excluding Joule heating.

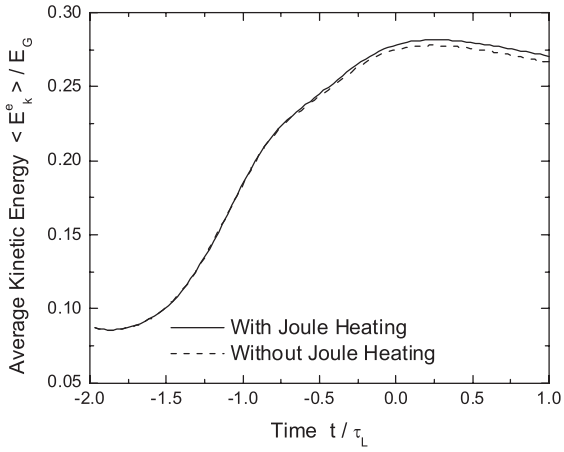

Figure 2. Average electron energy at scaled time of the laser pulse including and excluding Joule heating.

E. Conclusion

In our theoretical consideration, we have assumed that the wavelength of the laser radiation is longer than the other characteristic lengths so in the quantum-mechanical derivation we have ignored the spatial dependence of the electromagnetic field. In the initial quantum-mechanical calculations we have considered an electron-phonon system in a spatially uniform sinusoidal electric field of frequency Ω_L (dipole approximation). After the derivation of the kinetic equation for the electron energy distribution function,

the spatial dependence of the laser field is taken into account by explicitly including in the rates of the equation the spatially dependent envelope function that comprises the amplitude of the laser radiation electric field. The left hand side of the initially obtained kinetic equation is represented as a current in energy space. Consequently the energy dependent current is generalized to be energy and space dependent. The kinetic equation is augmented with spatially dependent currents and a generalized Fokker-Planck type equation in the energy-position manifold is obtained. Without invoking expressions for the electron vector mean-free-path related to the momentum relaxation and the electron mobility of the sample, from elementary arguments we have obtained the connection between the field dependent drift velocity in energy space and the drift velocity in real space.

II. QUANTUM KINETIC THEORY FOR SPATIO-TEMPORAL DYNAMICS OF CARRIERS IN LASER-EXCITED MATERIALS

A. Introduction

When dealing with a typical semiconductor-based optoelectronic device irradiated with laser light we consider the optical field, the created electron-hole plasma (EHP) and the crystal lattice of the chosen material. Light generation, propagation and amplification determine the behavior of the optical field. Carrier generation and recombination, electrical conduction and diffusion determine the behavior of the formed plasma. The photon energy of the laser field is converted and conserved as kinetic and thermal energy of the plasma and thermal energy of the lattice by creation and annihilation of phonons. All the described processes take place on different time and space scales but they should be treated in a self consistent manner with the appropriate coupled equations. Laser beam filamentation, dynamic beam steering, catastrophic optical damage, thermal lensing leading to formation of hot spots are cases requiring inclusion of spatial variation in the formalism describing the dynamics of optically generated carriers interacting with the phonons of the lattice. Inhomogeneous excitation, bulk filamentation laser damage, etc. lead to space dependent carrier distributions.

B. Theoretical model

When a spatially homogeneous system is excited by a spatially inhomogeneous laser field, the dynamical variables become inhomogeneous and off diagonal density matrices have to be introduced. A mixed momentum and real space representation is most similar to classical distribution function and is best suited for a comparison to semi-classical kinetics described by Boltzmann equation. A microscopic density matrix

theory is formulated accounting for arbitrary spatial inhomogeneities in the excitation conditions leading to space-dependent Boltzmann-Bloch transport equations for the description of spatio-temporal dynamics of electrons and holes of inhomogeneously excited materials such as semiconductors including the coherent interactions of carriers and the laser light field as well as transport due to spatial gradients and electrostatic forces. Only the classical character of the laser optical field is considered while accounting for the quantum mechanical properties of the semiconductor. Besides the interaction with the light field other important interactions occur in the semiconductor – Coulomb interaction among the carriers giving rise to screening and to thermalization of the nonequilibrium carrier distribution, as well as interaction with phonons leading to an energy exchange between the carriers and the crystal lattice. Based on typical length and time scales approximations are made with the aim of obtaining numerically tractable system of equations. We follow the approach in [11] but unlike them we treat all the scattering terms explicitly without resorting to relaxation time approximation [12]. We also include terms that lead to transitions between valence and conduction band – impact ionization and Auger recombination [3].

We consider a two-band model of an udoped semiconductor such as GaAs. In the laser-matter interaction process the physical variables that are directly related to observables of the system such as optical polarizations and distribution functions are all single-particle quantities calculated by the density matrix. To describe space-dependent phenomena a Wigner representation of the single-particle density matrix can be used. In Wigner representation the space-dependent distribution functions (intraband density matrices) of electrons and holes and polarization (interband density matrix) are defined as

$$f^e(\vec{k},\vec{r}) = \sum_{\vec{q}} e^{i\vec{q}\cdot\vec{r}}\langle c_{\vec{k}+\frac{1}{2}\vec{q}} + c_{\vec{k}-\frac{1}{2}\vec{q}}\rangle,\ f^h(\vec{k},\vec{r}) = \sum_{\vec{q}} e^{i\vec{q}\cdot\vec{r}}\langle d_{\vec{k}+\frac{1}{2}\vec{q}} + d_{\vec{k}-\frac{1}{2}\vec{q}}\rangle \text{ and}$$
$$p(\vec{k},\vec{r}) = \sum_{\vec{q}} e^{i\vec{q}\cdot\vec{r}}\langle d_{-\vec{k}+\frac{1}{2}\vec{q}} c_{\vec{k}+\frac{1}{2}\vec{q}}\rangle,$$

where $c^+_{\vec{k}}$ and $d^+_{\vec{k}}$($c_{\vec{k}}$ and $d_{\vec{k}}$) denote creation (annihilation) operators for electrons and holes with wave vector, respectively and the brackets denote the expectation value of these operators.

The single-particleHamiltonian describing the free carrier interacting with a classical light field as well as the free phonons is given by:

$$H_0 = \sum_{\vec{k}} \varepsilon^e_{\vec{k}} c^+_{\vec{k}} c_{\vec{k}} + \sum_{\vec{k}} \varepsilon^h_{\vec{k}} d^+_{\vec{k}} d_{\vec{k}} + \sum_{\vec{q}} \hbar\omega_{\vec{q}} b^+_{\vec{q}} b_{\vec{q}} -$$
$$- \sum_{\vec{k},\vec{q}} \left[\vec{\mu}_{cv}(\vec{k}) \cdot \vec{E}^+_{\vec{q}}(t) c^+_{\vec{k}+\frac{1}{2}\vec{q}} d^+_{-\vec{k}+\frac{1}{2}\vec{q}} + \vec{\mu}^*_{cv}(\vec{k}) \cdot \vec{E}^-_{\vec{q}}(t) d^+_{-\vec{k}+\frac{1}{2}\vec{q}} c^+_{\vec{k}+\frac{1}{2}\vec{q}} \right] \quad (1)$$

where $\mu\left(\vec{k}\right)$ is the component in the direction of the laser field polarization of the interband optical dipole matrix elememt between the electron state $\left|c,\vec{k}\right\rangle$ and hole state $\left|v,-\vec{k}\right\rangle$. The field is represented by two

counterpropagating waves and the positive frequency component is given by:

$$\vec{E}^{+}(\vec{r},t)=\frac{1}{2}\left(\vec{E}^{+}(\vec{r},t)\,e^{iK_z z-i\omega t}+\vec{E}^{-}(\vec{r},t)\,e^{-iK_z z-i\omega t}\right) \tag{2}$$

and is expanded in a Fourier series

$$\vec{E}^{+}(\vec{r},t)=\sum_{\vec{q}}\vec{E}^{0}_{\vec{q}}(t)\,e^{i(\vec{q}\cdot\vec{r}-\omega t)}=\sum_{\vec{q}}\vec{E}^{+}_{\vec{q}}(t)\,e^{i\vec{q}\cdot\vec{r}} \tag{3}$$

In the absence of an external light field the electron states are eigenstates of an ideal periodic lattice. Deviations from this idealized periodicity due to lattice vibrations lead to a coupling of the different electronic states. This interaction is described by the carrier-phonon Hamiltonian.

$$H_I^{cp}=\sum_{\vec{k},\vec{k}',\vec{q}}C_{\vec{q}}\left[c^{+}_{\vec{k}+\vec{q}}b_{\vec{q}}c_{\vec{k}}-c^{+}_{\vec{k}}b^{+}_{\vec{q}}c^{-}_{\vec{k}+\vec{q}}d_{\vec{k}+\vec{q}}+b_{\vec{q}}d^{+}_{\vec{k}}d_{\vec{k}}+b_{\vec{q}}+d_{\vec{k}+\vec{q}}\right], \tag{4}$$

where the electron- phonon coupling constant for interaction with optical phonons is:

$$\left|C_{\vec{q}}\right|^2=\left(\frac{\hbar\omega_{LO}}{2V}\right)\left[\frac{1}{\varepsilon_r(\infty)}-\frac{1}{\varepsilon_r(0)}\right]\left[\frac{e^2}{\varepsilon_0(q^2+Q_s^2)}\right] \tag{5}$$

$\varepsilon_r(\infty)$ and $\varepsilon_r(0)$ are the relative static and optical dielectric constant, respectively, ε_0 is the absolute dielectric constant of the vacuum, $\hbar\omega_{LO}$ is the optical phonon energy and V is the normalization volume. The charged carriers interact via the Coulomb potential $V_{\vec{q}}$ and the Hamiltonian describing carrier-carrier interaction processes conserving the number of particles per band is given by:

$$H_I^{cc}=\sum_{\vec{k},\vec{k}',\vec{q}}V_{\vec{q}}\left[\frac{1}{2}c^{+}_{\vec{k}}c^{+}_{\vec{k}'}c_{\vec{k}'+\vec{q}}c_{\vec{k}-\vec{q}}+\frac{1}{2}d^{+}_{\vec{k}}d^{+}_{\vec{k}'}d_{\vec{k}'+\vec{q}}d_{\vec{k}-\vec{q}}-c^{+}_{\vec{k}}d^{+}_{-\vec{k}'}d_{-\vec{k}'+\vec{q}}c_{\vec{k}-\vec{q}}\right] \tag{6}$$

This carrier-carrier Hamiltonian can be separated into a mean field (Haretree-Fock) H_{HF}^{cc} part and a remaining part depending on two-particle correlations H_{corr}^{cc}. The effective single particle Hamiltonian is $H_{eff}=H_0+H_{HF}^{cc}$. The correlation part of the carrier-carrier interaction Hamiltonian gives two phenomena: scattering processes between the carriers and screening of the bare Coulomb interaction.

The part of the perturbation Hamiltonian that yields impact ionization and its inverse process, Auger recombination is given by [3], [13]:

$$H_I^{cc(c-v)}=\sum_{\vec{k},\vec{k}',\vec{q}}\left[M_e(q)\,c^{+}_{\vec{k}+\vec{q}}c^{+}_{\vec{k}'-\vec{q}}d^{+}_{-\vec{k}'}c_{\vec{k}}+M_e^{*}(q)\,c^{+}_{\vec{k}}d_{-\vec{k}'}c_{\vec{k}'-\vec{q}}c^{+}_{\vec{k}+\vec{q}}\right]+$$
$$+\sum_{\vec{k},\vec{k}',\vec{q}}\left[M_h(q)\,d^{+}_{\vec{k}+\vec{q}}d^{+}_{\vec{k}'-\vec{q}}c^{+}_{-\vec{k}'}d_{\vec{k}}+M_h^{*}(q)\,d^{+}_{\vec{k}}c_{-\vec{k}'}d_{\vec{k}'-\vec{q}}d_{\vec{k}+\vec{q}}\right] \tag{7}$$

$M_e(q)=V_{\vec{q}}g_{\vec{q}}$, where $V_{\vec{q}}=\dfrac{4\pi e^2}{V\varepsilon_0 q^2}$ is the Coulomb potential and $g_{\vec{q}}$ is the interband-transition form factor.

C. Generalized Boltzmann-Bloch equations

By using Heisenberg's equations of motion, the equations of motion for the single-particle density matrices in Wigner representation can be derived. The effective single-particle Hamiltonian H_{eff} gives a closed set of equations for the distribution functions of electrons and holes and by interband polarization. Being Wigner distributions these quantities are functions of space and momentum but there is a big difference of time scales between the momentum space and real space dynamics. Scattering and dephasing processes lead to fast relaxation of the microscopic variables towards their local quasi-equilibrium values on a femtosecond time-scale while the spatial transport happens on a much slower time-scale (10 ps to ns). Because of the typical separation of time scales between the $\vec{k}$-space and $\vec{r}$-space dynamics, the influence of spatial gradients on the k-space dynamics is often negligible. However, some of the scattering terms in the equations of motion for the distribution functions conserve the density of carriers and therefore the density is not influenced by the fast relaxation processes and its spatial transport cannot be neglected. In the equation of motion for the polarization no conserved quantities exist and thus the spatial transport of polarization is usually not important. In principle the complete set of equations required is therefore the Maxwell-Boltzmann-Bloch-Poisson equations for the nonequilibrium distribution functions $f^{\alpha}\left(\vec{k},\vec{r}\right)$, interband polarization $p\left(\vec{k},\vec{r}\right)$, electric potential $\Phi\left(\vec{r}\right)$, and the laser field $\vec{\mathrm{E}}\left(\vec{r},t\right)$, with $\vec{k}$ and $\vec{r}$ being the two-dimensional (2D) vectors in reciprocal (momentum) space and real space, respectively.

Keeping the first order spatial derivatives of the distribution functions and neglecting any spatial transport of polarization, the equations of motion for electron and hole distribution functions are given by the generalized Boltzmann equations for two band model including the coherent interband transport contributions.

$$\frac{\partial}{\partial t}f^{\alpha}(\vec{k},\vec{r},t)+\frac{1}{\hbar}\frac{\partial\varepsilon^{\alpha}(\vec{k},\vec{r})}{\partial k}\cdot\frac{\partial f^{\alpha}(\vec{k},\vec{r},t)}{\partial r}-\frac{1}{\hbar}\frac{\partial}{\partial r}[\delta\varepsilon^{\alpha}(\vec{k},\vec{r})+q\Phi(r)]\cdot\frac{\partial f^{\alpha}(\vec{k},\vec{r},t)}{\partial k}= \\ =R^{\alpha}(\vec{k},\vec{r})+\frac{\partial}{\partial t}f^{\alpha}(\vec{k},\vec{r})_{col} \quad (8)$$

The lowest order contribution to the polarization is included, where the spatial coordinate enters only as a parameter and locally the dynamics coincide with those of the inhomogeneous case and there are no transport effects. This lowest order picture is sufficient to describe pump-probe experiments in which filamentation is observed.

$$\frac{\partial}{\partial t}p(\vec{k},\vec{r},t)=-\frac{i}{\hbar}[\varepsilon^{e}(\vec{k},\vec{r},t)+\varepsilon^{h}(-\vec{k},\vec{r},t)]p(\vec{k},\vec{r},t)- \\ -i\Omega(\vec{k},\vec{r})[f^{e}(\vec{k},\vec{r},t)+f^{h}(-\vec{k},\vec{r},t)-1]+\frac{\partial}{\partial t}p(\vec{k},\vec{r})_{col} \quad (9)$$

$$\varepsilon^{\alpha}\left(\vec{k},\vec{r}\right)=\varepsilon^{\alpha}\left(\vec{k}\right)+q^{\alpha}\Phi\left(r\right)+\delta\varepsilon^{\alpha}\left(\vec{k},\vec{r}\right),$$

$\varepsilon^{e,h}\left(\vec{k}\right) = \dfrac{\hbar^2 k^2}{2m_\alpha}$ is the single particle energy, $\delta\varepsilon^{\alpha}(\vec{k},\vec{r}) = -\sum_{\vec{k}'} f^{\alpha}(\vec{k},\vec{r})V^{s}_{\vec{k}'-\vec{k}} + \dfrac{1}{2}\sum_{\vec{k}'}[V^{s}_{\vec{k}'-\vec{k}} - V_{\vec{k}'-\vec{k}}]$ is the renormalization of the single particle carrier energy due to exchange interaction, $V^{s}_{\vec{q}} = \dfrac{V_{\vec{q}}}{\varepsilon(\vec{q},0)}$ is the screened Coulomb potential. The electrostatic potential due to the Hartree terms in the mean field Hamiltonian satisfies the Poisson equation:

$$\frac{\partial^2}{\partial r^2}\Phi(\vec{r}) = \frac{4\pi e}{\varepsilon_0 V}\sum_{\vec{k}}\left[f^{e}\left(\vec{k},\vec{r}\right) - f^{h}\left(\vec{k},\vec{r}\right)\right] \tag{10}$$

The generation rate in the equation (1) is given as follows:

$$R^{\alpha}\left(\vec{k},\vec{r}\right) = i\left[\Omega\left(\vec{k},\vec{r}\right)p*\left(\vec{k},\vec{r}\right) - \Omega*\left(\vec{k},\vec{r}\right)p\left(\vec{k},\vec{r}\right)\right], \tag{11}$$

where $\Omega\left(\vec{k},\vec{r}\right)$ is the renormalized Rabi frequency defined by:

$$\hbar\Omega\left(\vec{k},\vec{r}\right) = \mu\left(\vec{k}\right)\vec{\mathrm{E}}(\vec{r},t) + \sum_{\vec{k}'} p\left(\vec{k}',\vec{r}\right)V^{s}_{\vec{k}-\vec{k}'}. \tag{12}$$

The second term in the above expression is the internal field responsible for Coulomb enhancement.

D. Scattering processes

Within a semiclassical picture when scattering processes are described in terms of scattering rates, the scattering contributions to the equation of motion have the structure of the Boltzmann collision terms. The electron-phonon scattering rates are obtained from Fermi's golden rule and quadratic or higher order terms and terms involving simultaneous electron-phonon and hole-phonon interaction have been neglected in the polarization and in the carrier-phonon Hamiltonian. Incoherent scattering processes appear for the first time in second order contributions [3], [11], [14], [15].

Collisional contributions in equations (1) and (2) lead to relaxation in the carrier distributions and decay in the interband polarization:

$$\frac{\partial}{\partial t}f^{\alpha}\left(\vec{k},\vec{r}\right)_{col} = \frac{\partial}{\partial t}f^{\alpha}\left(\vec{k},\vec{r}\right)_{\alpha\alpha} + \frac{\partial}{\partial t}f^{\alpha}\left(\vec{k},\vec{r}\right)_{eh} + \frac{\partial}{\partial t}f^{\alpha}\left(\vec{k},\vec{r}\right)_{LO} \tag{13}$$

$$\frac{\partial}{\partial t}p\left(\vec{k},\vec{r}\right)_{col} = \sum_{\vec{q}}\left[W^{p}_{\vec{k},\vec{k}-\vec{q}}p\left(\vec{k}-\vec{q},\vec{r}\right) - W^{p}_{\vec{k}-\vec{q},\vec{k}}p\left(\vec{k},\vec{r}\right)\right] \tag{14}$$

The first term on the RHS of equation (3) depicts the scattering processes arising from the correlation part of the carrier-carrier Hamiltonian and the third term arises from the carrier-phonon Hamiltonian

$$\frac{\partial}{\partial t}f^{\alpha}(\vec{k},\vec{r})_{\alpha\alpha/LO} = \sum_{\vec{q}}[W^{e,h}_{\vec{k},\vec{k}-\vec{q}}f^{e,h}(\vec{k}-\vec{q},\vec{r})(1-f^{e,h}(\vec{k},\vec{r})) - W^{e,h}_{\vec{k}-\vec{q},\vec{k}}f^{e,h}(\vec{k},\vec{r})(1-f^{e,h}(\vec{k}-\vec{q},\vec{r}))] \tag{15}$$

where the scattering matrices are given by:

$$W^{e,h(\alpha\alpha)}_{\vec{k}-\vec{q},\vec{k}} = \frac{2\pi}{\hbar}\sum_{\alpha=e,h}\sum_{\vec{k}'}\left|V^s_{\vec{q}}\right|^2\delta\left(\varepsilon^{e,h}_{\vec{k}-\vec{q}}+\varepsilon^{\alpha}_{\vec{k}'+\vec{q}}-\varepsilon^{\alpha}_{\vec{k}'}-\varepsilon^{e,h}_{\vec{k}}\right)f^{\alpha}\left(\vec{k}',\vec{r}\right)\left[1-f^{\alpha}\left(\vec{k}'+\vec{q},\vec{r}\right)\right] \quad (16)$$

$$W^{e,h(LO)}_{\vec{k}-\vec{q},\vec{k}} = \frac{2\pi}{\hbar}\left|C^{e,h}_{q}\right|^2(N_q+1)\,\delta\left(\varepsilon^{e,h}_{\vec{k}-\vec{q}}-\varepsilon^{e,h}_{\vec{k}}+\hbar\omega_q\right)+\frac{2\pi}{\hbar}\left|C^{e,h}_{q}\right|^2 N_q\delta\left(\varepsilon^{e,h}_{\vec{k}-\vec{q}}-\varepsilon^{e,h}_{\vec{k}}-\hbar\omega_q\right) \quad (17)$$

The Boltzmann scattering matrices $W^{e,h}_{\vec{k}-\vec{q},\vec{k}}$ for electrons and holes are real quantities and the scattering matices $W^{p}_{\vec{k}-\vec{q},\vec{k}}$ in the equation of motion for the polarization are complex and the real part is related to $W^{e,h}_{\vec{k}-\vec{q},\vec{k}}$ according to:

$$\begin{aligned}\mathrm{Re}\left(W^{p}_{\vec{k}-\vec{q},\vec{k}}\right) = \frac{1}{2}W^{e}_{\vec{k}-\vec{q},\vec{k}}\left[1-f^{e}\left(\vec{k}-\vec{q},\vec{r}\right)\right]+W^{e}_{\vec{k},\vec{k}-\vec{q}}f^{e}\left(\vec{k}-\vec{q},\vec{r}\right)+\\ +\frac{1}{2}W^{h}_{\vec{q}-\vec{k},-\vec{k}}\left[1-f^{h}\left(\vec{q}-\vec{k},\vec{r}\right)\right]+W^{h}_{-\vec{k},\vec{q}-\vec{k}}f^{h}\left(\vec{q}-\vec{k},\vec{r}\right) \quad (18)\end{aligned}$$

The real part of $W^{p}_{\vec{k}-\vec{q},\vec{k}}$ describes scattering processes leading to a dephasing of the polarization and the imaginary part describes second-order contributions to the band-gap renormalization.

The carrier-carrier scattering rate in the collisional contribution to the polarization equation describing the effect of correlations is given by:

$$\begin{aligned}W^{p(\alpha\alpha)}_{\vec{k}-\vec{q},\vec{k}} = \frac{\pi}{\hbar}\sum_{\alpha=e,h}\sum_{\vec{k}'}\left|V^s_{\vec{q}}\right|^2\delta\left(\varepsilon^{e,h}_{\vec{k}-\vec{q}}+\varepsilon^{\alpha}_{\vec{k}'+\vec{q}}-\varepsilon^{\alpha}_{\vec{k}'}-\varepsilon^{e,h}_{\vec{k}}\right)\times\\ \times f^{\alpha}\left(\vec{k}',\vec{r}\right)\left[1-f^{\alpha}\left(\vec{k}'+\vec{q},\vec{r}\right)\right]\times\left[1-f^{e,h}\left(\vec{k}-\vec{q},\vec{r}\right)\right]+\\ +f^{e,h}\left(\vec{k}-\vec{q},\vec{r}\right)f^{\alpha}\left(\vec{k}'+\vec{q},\vec{r}\right)\left[1-f^{\alpha}\left(\vec{k}'+\vec{q},\vec{r}\right)\right] \quad (19)\end{aligned}$$

The carrier-phonon scattering rate in the collisional contribution to the polarization equation is given by:

$$\begin{aligned}W^{p(LO)}_{\vec{k}-\vec{q},\vec{k}} = \frac{\pi}{\hbar}\left|C_q\right|^2\delta\left(\varepsilon^{e,h}_{\vec{k}-\vec{q}}-\varepsilon^{e,h}_{\vec{k}}+\hbar\omega_q\right)\left\{N_q f^{e,h}\left(\vec{k}-\vec{q},\vec{r}\right)+(N_q+1)\left[1-f^{e,h}\left(\vec{k}-\vec{q},\vec{r}\right)\right]\right\}+\\ +\frac{\pi}{\hbar}\left|C_q\right|^2\delta\left(\varepsilon^{e,h}_{\vec{k}-\vec{q}}-\varepsilon^{e,h}_{\vec{k}}-\hbar\omega_q\right)\left\{(N_q+1)\,f^{e,h}\left(\vec{k}-\vec{q},\vec{r}\right)+N_q\left[1-f^{e,h}\left(\vec{k}-\vec{q},\vec{r}\right)\right]\right\} \quad (20)\end{aligned}$$

E. Impact ionization and Auger recombination

Since we interested in the processes of laser damage and filamentation in the semiconductor material, we include the $\frac{\partial}{\partial t}f^{\alpha}\left(\vec{k},\vec{r}\right)_{eh}$ terms that lead to transitions between valence and conduction bands, i.e. impact ionization term and Auger recombination term [3]. Impact ionization and Auger recombination are second-order two-particle Coulomb scattering processes (proportional to Coulomb scattering). In a case when we have a homogeneous system (material) that is either homogeneously or inhomogeneously excited the Coulomb matrix elements depend on the momentum transfer $\vec{q}$ only, i.e. $\propto \left|V_{\vec{q}}\right|^2$.

These contributions are derived using second-order perturbation theory such as Coulomb scattering [3] which is very important to conduction-electron dynamics and the change of electron density.

$$\begin{aligned}\frac{\partial}{\partial t}f^e\left(\vec{k},\vec{r}\right)_{e-h(imp)} =& N_{eff}\frac{2\pi}{\hbar}\sum_{\vec{q}}\left|V_{\vec{q}}\right|^2 g_{\vec{q}}\delta\left(2\varepsilon^e_{\vec{k}}-\varepsilon^e_{\left|\vec{k}+\vec{q}\right|}+\varepsilon^h_{\left|\vec{k}-\vec{q}\right|}+E_G\right)\times \\ &\times\left[1-f^e\left(\vec{k},\vec{r}\right)\right]^2 f^e\left(\vec{k}+\vec{q},\vec{r}\right)\left[1-f^h\left(\vec{k}-\vec{q},\vec{r}\right)\right]+ \\ &+N_{eff}\frac{2\pi}{\hbar}\sum_{\vec{q}}\left|V_{\vec{q}}\right|^2 g_{\vec{q}}\delta\left(2\varepsilon^e_{\left|\vec{k}-\vec{q}\right|}-\varepsilon^e_{\vec{k}}+\varepsilon^h_{\left|\vec{k}-2\vec{q}\right|}+E_G\right)\times \\ &\times f^e\left(\vec{k},\vec{r}\right)\left[1-f^e\left(\vec{k}-\vec{q},\vec{r}\right)\right]^2\left[1-f^h\left(\vec{k}-2\vec{q},\vec{r}\right)\right] \end{aligned} \tag{21}$$

$$\begin{aligned}\frac{\partial}{\partial t}f^e\left(\vec{k},\vec{r}\right)_{e-h(rec)} =& \frac{2\pi}{\hbar}\sum_{\vec{q}}\left|V_{\vec{q}}\right|^2 g_{\vec{q}}\delta\left(\varepsilon^e_{\vec{k}}-2\varepsilon^e_{\vec{k}-\vec{q}}-\varepsilon^h_{\vec{k}-2\vec{q}}-E_G\right)\times \\ &\times\left[1-f^e\left(\vec{k},\vec{r}\right)\right]\times\left[f^e\left(\vec{k}-\vec{q},\vec{r}\right)\right]^2 f^h\left(\vec{k}-2\vec{q},\vec{r}\right)+ \\ &+\frac{2\pi}{\hbar}\sum_{\vec{q}}\left|V_{\vec{q}}\right|^2 g_{\vec{q}}\delta\left(\varepsilon^e_{\vec{k}+\vec{q}}-2\varepsilon^e_{\vec{k}}-\varepsilon^h_{\vec{k}-\vec{q}}-E_G\right)\times \\ &\times\left[f^e\left(\vec{k},\vec{r}\right)\right]^2\left[1-f^e\left(\vec{k}+\vec{q},\vec{r}\right)\right]f^h\left(\vec{k}-\vec{q},\vec{r}\right) \end{aligned} \tag{22}$$

with $g_{\vec{q}}\approx 2\left(m^*_e/m_0\right)$.

The semiclassical generation rate for carrier-light interaction is obtained by eliminating the polarization as independent variable [15]. This is done by solving the equation for polarization within the adiabatic and Markov approximation. For Gaussian pulse with a space dependent amplitude

$$E\left(\vec{r},t\right)=E_L\left(\vec{r}\right)\exp\left[-\left(t/\tau_L\right)^2\right] \tag{23}$$

the time integration of the polarization equation (2) gives

$$R^\alpha\left(\vec{k},\vec{r}\right)=\left(2\pi\right)^{1/2}\left(\frac{M_k E_L\left(\vec{r}\right)}{\hbar}\right)^2\tau_L\exp\left[-2\left(t/\tau_L\right)^2\right]\exp\left[-\frac{1}{2}\left(\tau_L\Delta\omega_k\right)^2\right]\times\left[1-f^e\left(\vec{k},\vec{r}\right)-f^h\left(\vec{k},\vec{r}\right)\right] \tag{24}$$

where

$$\Delta\omega_k=\omega_L-\frac{1}{\hbar}\left(\varepsilon^e\left(\vec{k},\vec{r}\right)+\varepsilon^h\left(\vec{k},\vec{r}\right)\right) \tag{25}$$

is the detuning of a given transition with wavevector $\vec{k}$ from resonance, ω_L being the laser frequency.

The time integration is possible only under the assumption that during the laser pulse the polarization is not influenced by any scattering processes leading to phase-breaking during carrier generation. The only density dependence of this rate is due to phase-space filling. While adiabatic elimination of polarization leads to a simple closure of the total set of equations, this set of equations have a severe deficiency, especially

in the presence of any kind of spatial inhomogeneity. That is why in the formulated approach we are keeping the Boltznann-Bloch transport equations for the three distribution functions $f^e\left(\vec{k},\vec{r}\right)$, $f^h\left(\vec{k},\vec{r}\right)$, $p\left(\vec{k},\vec{r}\right)$, though all transport terms involving explicit spatial variation of $p\left(\vec{k},\vec{r}\right)$ are ignored. For ultrafast spatial inhomogeneous processes such spatial terms in the polarization equations should be important.

F. Numerical procedures in progress

The microscopic dynamics of the distribution unction and the nonlinear polarization are governed by the equations of motion (1) and (2). The $\vec{k}$-resolved interband polarization equations have to be solved self-consistently for all space and time grid points. A full kinetic treatment of the scattering processes will be performed by using e.g. Monte Carlo simulations. Generalized Monte Carlo methods taking into account phase relations between different type of carriers (polarization effects), interaction of carriers with external coherent inhomogeneous electromagnetic field (generation effects), and the correlation and renormalization effects associated with carrier-carrier interaction can be utilized [16].

G. Conclusion

A microscopic quantum-kinetic theory based on density matrix formalism [17] is formulated to describe the processes of short pulse laser interaction with materials such as semiconductors accounting for arbitrary spatial inhomogeneities in the excitation conditions and other spatial phenomena such as filamentation of tightly focused femtosecond laser pulses, structural modification and catastrophic optical damage. A system of Boltzmann-Bloch transport equations are established that include both space and momentum dependence of the electron and hole distribution functions and the polarization. Microscopic electron-phonon and electron-electron scattering terms as well as scattering terms that lead to transitions between valence and conduction bands, i.e. impact ionization and recombination terms, are included explicitly in the equations. The formulated theory describes the spatio-temporal dynamics of electrons and holes in inhomogeneously excited materials including the coherent interactions of carriers and the laser light field as well as transport due to spatial gradients and electrostatic forces.

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Symbols, Abbreviations, and Acronyms are explained in the text