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Quantum Kinetic Approach for Spatio-Temporal Carrier Dynamics in Femtosecond-Laser Irradiated Materials

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Abstract. A microscopic quantum-kinetic theory based on density matrix approach (using Wigner function representation) 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 is established that includes 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 carrier dynamics 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.

1 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.

2 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 [1] but unlike them we treat all the scattering terms explicitly without resorting to relaxation time approximation [2]. 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 undoped 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

$$f^e(\vec{k},\vec{r}) = \sum_{\vec{q}} e^{i\vec{q}\cdot\vec{r}} \langle c^{\dagger}_{\vec{k}+\frac{1}{2}\vec{q}} c_{\vec{k}-\frac{1}{2}\vec{q}} \rangle,$$

$$f^h(\vec{k},\vec{r}) = \sum_{\vec{q}} e^{i\vec{q}\cdot\vec{r}} \langle d^{\dagger}_{\vec{k}+\frac{1}{2}\vec{q}} d_{\vec{k}-\frac{1}{2}\vec{q}} \rangle$$

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 $c^{\dagger}_{\vec{k}}$ and $d^{\dagger}_{\vec{k}}$($c_{\vec{k}}$ 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:

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

where $\mu(\vec{k})$ is the component in the direction of the laser field polarization of the interband optical dipole matrix element between the electron state $|c, \vec{k}\rangle$ and hole state $|v, -\vec{k}\rangle$. The field is represented by two counterpropagating waves and the positive frequency component is given by:

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

and is expanded in a Fourier series

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

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.

$$H^{cp}_I = \sum_{\vec{k},\vec{k}',\vec{q}} C_{\vec{q}} \left[c^\dagger_{\vec{k}+\vec{q}} b_{\vec{q}} c_{\vec{k}} - c^\dagger_{\vec{k}} b^\dagger_{\vec{q}} c_{\vec{k}+\vec{q}} - d^\dagger_{\vec{k}+\vec{q}} b_{\vec{q}} d_{\vec{k}} + d^\dagger_{\vec{k}} b^\dagger_{\vec{q}} d_{\vec{k}+\vec{q}} \right], \quad (4)$$

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

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

$\varepsilon_r(\infty)$ 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:

$$H^{cc}_I = \sum_{\vec{k},\vec{k}',\vec{q}} V_{\vec{q}} \left[\frac{1}{2} c^\dagger_{\vec{k}} c^\dagger_{\vec{k}'} c_{\vec{k}'+\vec{q}} c_{\vec{k}-\vec{q}} + \frac{1}{2} d^\dagger_{\vec{k}} d^\dagger_{\vec{k}'} d_{\vec{k}'+\vec{q}} d_{\vec{k}-\vec{q}} - c^\dagger_{\vec{k}} d^\dagger_{-\vec{k}'} d_{-\vec{k}'+\vec{q}} c_{\vec{k}-\vec{q}} \right] \quad (6)$$

This carrier-carrier Hamiltonian can be separated into a mean field (Haretree-Fock) H^{cc}_{HF} 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], [4]:

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

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

3 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 $f^\alpha(\vec{k},\vec{r})$, interband polarization $p(\vec{k},\vec{r})$, electric potential $\Phi(\vec{r})$, and the laser field $\vec{\mathrm{E}}(\vec{r},t)$, 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, $V^{s}_{\vec{q}} = \dfrac{V_{\vec{q}}}{\varepsilon\left(\vec{q},0\right)}$ is the screened Coulomb potential. The electrostatic potential due to the Hartree terms in the mean field Hamiltonian satisfies the Poisson equation:

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

The generation rate in the equation (8) is given as follows:

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

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

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

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

4 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 [1,3,5,6].

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

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

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

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

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

where the scattering matrices are given by:

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

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

The Boltzmann scattering matrices $W^{e,h}_{\vec{k}-\vec{q},\vec{k}}$ for electrons and holes are real quantities and the scattering matrices $W^{p}_{\vec{k}-\vec{q},\vec{k}}$ 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:

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

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:

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

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

$$\begin{aligned}W^{p(LO)}_{\vec{k}-\vec{q},\vec{k}} &= \frac{\pi}{\hbar}\left|C_{q}\right|^{2}\delta\left(\varepsilon^{e,h}_{\vec{k}-\vec{q}}-\varepsilon^{e,h}_{\vec{k}}+\hbar\omega_{q}\right)\left\{N_{q}f^{e,h}\left(\vec{k}-\vec{q},\vec{r}\right)\right.\\ &+\left.\left(N_{q}+1\right)\left[1-f^{e,h}\left(\vec{k}-\vec{q},\vec{r}\right)\right]\right\}+\frac{\pi}{\hbar}\left|C_{q}\right|^{2}\delta\left(\varepsilon^{e,h}_{\vec{k}-\vec{q}}-\varepsilon^{e,h}_{\vec{k}}-\hbar\omega_{q}\right)\\ &\times\left\{\left(N_{q}+1\right)f^{e,h}\left(\vec{k}-\vec{q},\vec{r}\right)+N_{q}\left[1-f^{e,h}\left(\vec{k}-\vec{q},\vec{r}\right)\right]\right\} \qquad (20)\end{aligned}$$

5 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.

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

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

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 [6]. 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

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

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 $f^e(\vec{k}, \vec{r})$, $f^h(\vec{k}, \vec{r})$, $p(\vec{k}, \vec{r})$, though all transport terms involving explicit spatial variation of $p(\vec{k}, \vec{r})$ are ignored. For ultrafast spatial inhomogeneous processes such spatial terms in the polarization equations should be important.

6 Numerical Procedures in Progress

The microscopic dynamics of the distribution function 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 [7].

7 Conclusion

A microscopic quantum-kinetic theory based on density matrix formalism [8] 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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