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Spatio-temporal carrier dynamics in ultrashort-pulse laser irradiated materials

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ABSTRACT

A space and time-dependent quantum-kinetic theory has been formulated based on previous theoretical approaches to study the spatio-temporal microscopic carrier dynamics in laser excited semiconductor materials that accounts for the effects of inhomogeneous excitation and structural inhomogeneities due to bulk filamentation damage and micro/nano structuring. The approach involves extensive computational effort.

1. Introduction

When dealing with a typical semiconductor-based optoelectronic device irradiated with laser light we consider the created electron–hole plasma (EHP) in the crystal lattice of the chosen material. 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. Inhomogeneous excitation, bulk filamentation laser damage, etc. require inclusion of spatial variation in the formalism describing the dynamics of optically generated carriers and lead to space-dependent carrier distributions.

2. Theoretical model

When a spatially homogeneous system is excited by a spatially inhomogeneous laser field or a spatially dependent filamentary damage is induced, 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 semiclassical kinetics described by Boltzmann equation. By using mean-field approximation to the correlation of the electron and hole operators and dipole-coupling approximation to the interaction with the external electromagnetic field, an effective Hamiltonian is obtained in terms of the ascending–descending operators. On the basis of the effective Hamiltonian,the Boltzmann–Bloch equations for the description of spatio-temporal dynamics of electrons and holes of inhomogeneously excited semiconductors including the coherent interactions of carriers and the laser light field as well as transport due to spatial gradients and electrostatic forces are obtained. Besides the interaction with the light field other important interactions in the semiconductor such as 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 are included. *We follow the approach in Ref.* [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 i.e. 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

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

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.

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

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

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 the hole state $|v,-\vec{k}\rangle$. The field is represented by two counterpropagating waves and the positive frequency component is given by

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

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.

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

where $C_{\vec{q}}$ is the electron–phonon coupling constant for interaction with optical phonons, $\varepsilon_r(\infty)$ 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:

$$H^{cc}_{I}=\sum_{\vec{k},\vec{k}',\vec{q}}V_{\vec{q}}\left[\frac{1}{2}c^{\dagger}_{\vec{k}}c^{\dagger}_{\vec{k}'}c_{\vec{k}'+\vec{q}}c_{\vec{k}-\vec{q}}+\frac{1}{2}d^{\dagger}_{\vec{k}}d^{\dagger}_{\vec{k}'}d_{\vec{k}'+\vec{q}}d_{\vec{k}-\vec{q}}-c^{\dagger}_{\vec{k}}d^{\dagger}_{-\vec{k}'}d_{-\vec{k}'+\vec{q}}c_{\vec{k}-\vec{q}}\right]. \quad (5)$$

The 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^{cc}_{corr}. The effective single-particle Hamiltonian is $H_{eff}=H_0+H^{cc}_{HF}$. The correlation part of the carrier–carrier interaction Hamiltonian gives two phenomena: scattering processes between the carriers and the 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]

$$H^{cc(c-v)}_{I}=\sum_{\vec{k},\vec{k}',\vec{q}}[M_e(q)c^{\dagger}_{\vec{k}+\vec{q}}c^{\dagger}_{\vec{k}'-\vec{q}}d^{\dagger}_{-\vec{k}'}c_{\vec{k}}+M^{*}_{e}(q)c^{\dagger}_{\vec{k}}d_{-\vec{k}'}c_{\vec{k}'-\vec{q}}c^{\dagger}_{\vec{k}+\vec{q}}]$$
$$+\sum_{\vec{k},\vec{k}',\vec{q}}[M_h(q)d^{\dagger}_{\vec{k}+\vec{q}}d^{\dagger}_{\vec{k}'-\vec{q}}c^{\dagger}_{-\vec{k}'}d_{\vec{k}}+M^{*}_{h}(q)d^{\dagger}_{\vec{k}}c_{-\vec{k}'}d_{\vec{k}'-\vec{q}}d_{\vec{k}+\vec{q}}] \quad (6)$$

$M_e(q)=V_{\vec{q}}g_{\vec{q}}$, where $V_{\vec{q}}=e^2/V\varepsilon_0\varepsilon_r q^2$ 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 for 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 the 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 a 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 $\vec{E}(\vec{r},t)$, 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 contributions.

$$\frac{\partial}{\partial t}f^{\alpha}(\vec{k},\vec{r},t)+\frac{1}{\hbar}\frac{\partial\varepsilon^{\alpha}(\vec{k},\vec{r})}{\partial k}\cdot\frac{\partial f^{\alpha}(\vec{k},\vec{r},t)}{\partial r}-\frac{1}{\hbar}\frac{\partial}{\partial r}[\delta\varepsilon^{\alpha}(\vec{k},\vec{r})+q\Phi(r)]\cdot\frac{\partial f^{\alpha}(\vec{k},\vec{r},t)}{\partial k}$$
$$=R^{\alpha}(\vec{k},\vec{r})+\frac{\partial}{\partial t}f^{\alpha}(\vec{k},\vec{r})_{col}. \quad (7)$$

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.

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

$$\varepsilon^{\alpha}(\vec{k},\vec{r})=\varepsilon^{\alpha}(\vec{k})+q^{\alpha}\Phi(r)+\delta\varepsilon^{\alpha}(\vec{k},\vec{r}) \quad (8)$$

$\varepsilon^{e,h}(\vec{k})=\hbar^2k^2/2m_{\alpha}$ is the single-particle energy,

$$\delta\varepsilon^{\alpha}(\vec{k},\vec{r})=-\sum_{\vec{k}'}f^{\alpha}(\vec{k},\vec{r})V^{s}_{\vec{k}'-\vec{k}}+\frac{1}{2}\sum_{\vec{k}'}[V^{s}_{\vec{k}'-\vec{k}}-V_{\vec{k}'-\vec{k}}]$$

is the renormalization of the single-particle carrier energy due to exchange interaction, $V^{s}_{\vec{q}}$ is the screened Coulomb potential. The electrostatic potential due to the Hartree terms in the mean field Hamiltonian satisfies the Poisson equation.

The generation rate in the Eq. (7) is given as follows:

$$R^{\alpha}(\vec{k},\vec{r})=i[\Omega(\vec{k},\vec{r})p^{*}(\vec{k},\vec{r})-\Omega^{*}(\vec{k},\vec{r})p(\vec{k},\vec{r})] \quad (9)$$

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

$$\hbar\Omega(\vec{k},\vec{r})=\mu(\vec{k})\vec{E}(\vec{r},t)+\sum_{\vec{k}'}p(\vec{k}',\vec{r})V^{s}_{\vec{k}-\vec{k}'}. \quad (10)$$

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