Non-local quantum–kinetic equation for laser-excited semiconductors

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Abstract. A generalized non-local Fokker-Planck type kinetic equation obtained on the basis of quantum-mechanical formalism is used to study the conduction electron dynamics in model semiconductors excited by spatially inhomogeneous pulsed laser irradiation. In this approach the electrons are assumed to undergo a drift-diffusion motion in the energy-position manifold.

1. Introduction
Ultra-short laser-induced breakdown studies have revealed the possibility of self-channeling ultrafast light in condensed media with the strong involvement of photon 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 \( \tilde{E} \) of the electric field of the laser

\[
\tilde{E}(\vec{r},t) = \frac{\Omega_L \mu_0}{2k_0} \tilde{E}(\vec{r},t) e^{i(k_0 r - \Omega_L t)} + c.c. \quad (\text{for a pulsed beam centered on the operating frequency } \Omega_L, \text{ wave number } k_0)
\]

where the electromagnetic radiation is polarized in the \( x \) direction \( \tilde{E} = (E,0,0) \). The free electrons motion is governed by the oscillation of the inhomogeneous optical field beating at \( \Omega_L \) 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 \( \nabla n \) 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 generalize the kinetic equation for the electron energy distribution function to obtain a multivariate Fokker-Planck kinetic equation accounting for the spatial dependence of electron transport. The rates in the equation explicitly include the effect of laser field energy absorption by the free electrons and the joule heating effect.

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2. Theoretical picture

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 $1 \ll \Omega_L \tau_p \ll \sqrt{\varepsilon}$ (the electromagnetic field frequency exceeds appreciably the frequency of electron-phonon collisions), $\Omega >> \omega_p$, $\bar{\varepsilon} \tau_p >> 1$ (where $\tau_p$ is the momentum relaxation time, $\omega_p$ is the electron plasma frequency and $\bar{\varepsilon}$ and $\bar{\nu}$ 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 $A(t) = A \sin(\Omega_L t)$ and find the solution of the Schrödinger equation

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

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 $\hbar \omega_k \ll E_s^*$, where $E_s^* = \hbar k_e^2 / 2m_e^*$ and $\hbar \omega_k$ 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 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:

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

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

$$V(E, t) = V_T(E) + V_p(E) + \frac{1}{3} \sigma(\Omega_L) E^2 + A_T(E) \frac{\tau_p}{3} \sigma(\Omega_L) E^2,$$

$$D(E, t) = D_T(E) + D_p(E) + \frac{2}{3} \sigma(\Omega_L) E^2 E_k^e + V_T(E) \frac{\tau_p}{3} \sigma(\Omega_L) E^2,$$

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

\[
V_F(E_k) = \frac{2\pi}{\hbar} \sum_{\nu \lambda} |C_{\nu \lambda}|^2 \hbar \omega_{\nu \lambda} \times \left[ \left(N^{ph}_{\nu \lambda} \delta(E_k - E_{\nu + \lambda} - \hbar \omega_{\nu \lambda}) - (N^{ph}_{\nu \lambda} + 1) \delta(E_k - E_{\nu + \lambda} + \hbar \omega_{\nu \lambda}) \right) \right],
\]

(5)

\[
V_F = \frac{2\pi}{\hbar} \sum_{\nu \lambda} |C_{\nu \lambda}|^2 \hbar \omega_{\nu \lambda} \times \left[ \left(N^{ph}_{\nu \lambda} \delta(E_k - E_{\nu + \lambda} - \hbar \omega_{\nu \lambda} + \hOmega) + \delta(E_k - E_{\nu + \lambda} - \hbar \omega_{\nu \lambda} - \hOmega) \right) + (N^{ph}_{\nu \lambda} + 1) \delta(E_k - E_{\nu + \lambda} + \hbar \omega_{\nu \lambda}) \right],
\]

(6)

\[
D_T(E_k) = \frac{\pi}{\hbar} \sum_{\nu \lambda} |C_{\nu \lambda}|^2 \hbar \omega_{\nu \lambda} \times \left[ \left(N^{ph}_{\nu \lambda} \delta(E_k - E_{\nu + \lambda} - \hbar \omega_{\nu \lambda}) - (N^{ph}_{\nu \lambda} + 1) \delta(E_k - E_{\nu + \lambda} + \hbar \omega_{\nu \lambda}) \right) \right],
\]

(7)

\[
D_F = \frac{\pi}{4\hbar} \sum_{\nu \lambda} |C_{\nu \lambda}|^2 \hbar \omega_{\nu \lambda} \times \left[ \left(N^{ph}_{\nu \lambda} \delta(E_k - E_{\nu + \lambda} - \hbar \omega_{\nu \lambda} + \hOmega) + \delta(E_k - E_{\nu + \lambda} - \hbar \omega_{\nu \lambda} - \hOmega) \right) + (N^{ph}_{\nu \lambda} + 1) \delta(E_k - E_{\nu + \lambda} + \hbar \omega_{\nu \lambda}) \right],
\]

(8)

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

\[
A_T(E_k) = \frac{2\pi}{\hbar} \sum_{\nu \lambda} |C_{\nu \lambda}|^2 \hbar \omega_{\nu \lambda} \times \left[ \left(N^{ph}_{\nu \lambda} \delta(E_k - E_{\nu + \lambda} - \hbar \omega_{\nu \lambda}) - (N^{ph}_{\nu \lambda} + 1) \delta(E_k - E_{\nu + \lambda} + \hbar \omega_{\nu \lambda}) \right) \right],
\]

(9)

\[
A_F = \frac{\pi}{2\hbar} \sum_{\nu \lambda} |C_{\nu \lambda}|^2 \hbar \omega_{\nu \lambda} \times \left[ \left(N^{ph}_{\nu \lambda} \delta(E_k - E_{\nu + \lambda} - \hbar \omega_{\nu \lambda} + \hOmega) + \delta(E_k - E_{\nu + \lambda} - \hbar \omega_{\nu \lambda} - \hOmega) \right) + (N^{ph}_{\nu \lambda} + 1) \delta(E_k - E_{\nu + \lambda} + \hbar \omega_{\nu \lambda}) \right],
\]

(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, \(|C_{\nu \lambda}|\) is the electron-phonon coupling and \(\mathcal{Z}_{\nu \lambda}(\epsilon, \bar{E}) = \left(\epsilon, \bar{E}(t) \right)/m^* \Omega^2_{\nu \lambda}\). 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.

3. Generalized Fokker-Planck kinetic equation

By explicitly substituting the spatially dependent envelope of the laser \(\bar{E}(\bar{r}, t)\) field in equations (3) - (10) the space and energy dependent rates \(V(\bar{r}, E, t)\) and \(D(\bar{r}, E, t)\) are obtained. Equation (2) is different from the standard Fokker-Planck equation since it cannot be written as a conservation law for particle number and current in energy space even in the absence source terms on the right hand side. Defining a current in energy space:

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

equation (2) can be written as
\[
\frac{\partial f^e(E,t)}{\partial t} + \frac{\partial f^e(E,t)}{\partial E} + 2 \frac{\partial D(E,t)}{\partial E} \frac{\partial f^e(E,t)}{\partial E} = A(E,t)f^e(E,t) + \tilde{A}(E,t)f^e(E,t) + S(E,t),
\]

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

For the time being we ignore the third term on the left-hand side of equation (11). The Joule heating and diffusion rates of equation (2) both contain expressions proportional to the electric field envelope and could be thus generalized to include space dependence. The electron distribution function is also generalized to include space dependence. Then a generalized current density term is defined reflecting the electron energy gain or loss and electron energy diffusion:

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

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] we generalize equation (11) to a continuity equation in the energy-position manifold to describe electron transport in an inhomogeneous laser field by continuous energy and coordinate changes

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

where the Greek indices run from 1 to 3.

Due to our choice of laser field polarization in the \( x \) direction, we get drift-diffusion equations in \((E,\vec{r})\) space:

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

\[
J^y(E,\vec{r},t) = -\frac{\partial [eE(\vec{r},t)D_x(E,\vec{r},t)f^e(E,\vec{r},t)]}{\partial E} - \frac{\partial [D_y(E,\vec{r},t)f^e(E,\vec{r},t)]}{\partial y},
\]

\[
J^z(E,\vec{r},t) = -\frac{\partial [eE(\vec{r},t)D_x(E,\vec{r},t)f^e(E,\vec{r},t)]}{\partial E} - \frac{\partial [D_z(E,\vec{r},t)f^e(E,\vec{r},t)]}{\partial z}.
\]

In equations (13)-(15), \( E(\vec{r},t) \) is the laser field envelope. We explicitly substitute the spatially dependent envelope of the laser field in 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.

4. 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 quantum mechanically derived field dependent velocity in energy space. An electron with momentum $\vec{p}_0$ and energy $E(\vec{p}_0)$ at time $t = 0$ is considered. Defining $\vec{V}_F = e \vec{E} \cdot \vec{V}_p$, where $\vec{E}$ denotes the laser electric field, the energy change before any collision occurs is: $E(t) - E_0 = (\vec{\partial}_F E)_{p_0} t + 1/2 (\vec{\partial}_F^2 E)_{p_0} t^2 + \ldots$ The probability for the electron starting from $\vec{p}_0$ not to experience collisions between 0 and $t$ is $\exp\left[-\int_0^t \text{d}t/[\tau(\vec{p})]\right]$, where account is taken of the change in $\tau$ as the electron momentum changes in time $d\vec{p}/dt = e\vec{E}$. Upon first order expansion of $\tau(\vec{p}_0 + e\vec{E}t)$, we obtain $c(t) = \exp\left[-t/\tau_0\right]\left[1 + 1/2(t/\tau_0)^2 (\vec{\partial}_F \tau)_{p_0}\right] + O(\vec{E}^2)$, where $\tau_0 = \tau(\vec{p}_0)$.

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

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

Then the average energy gained over a collisionless flight (of order $\vec{E}^2$) is:

$$\langle E(t) - E_0 \rangle = (\vec{\partial}_F E)_{p_0} t + (\vec{\partial}_F^2 E)_{p_0} t^2 + \ldots$$

so $\langle E(t) - E_0 \rangle = e\vec{E} \cdot \vec{V}_p (\vec{p}_0) + (\vec{\partial}_F \vec{V}_p)_{p_0} \|\vec{p}_0\|$, and

$$V_F(E_0) = \left(\frac{dE}{dt}\right)_F = e\vec{E} \cdot \left[\vec{V}_g(\vec{p}_0) + (\vec{\partial}_F \vec{V}_g)_{p_0}\right] \|\vec{p}_0\|,$$

with $\vec{V}_d(E_0) = \int_{E_0} \vec{V}_g(\vec{p}_0) dS_p/\|\vec{V}_g\|$, where $dS_p/\|\vec{V}_g\|$ are the number of states in a surface element $dS_p$ in momentum space. From the above expressions the relation between the field dependent drift velocity and the drift velocity in coordinate space is obtained: $V_F(E_0) = e\vec{E} \cdot \vec{V}_d(E_0)$ and for the choice of laser field polarization

$$v_{ds}(E, \vec{r}, t) = \frac{V_F(E, \vec{r}, t)}{(e\vec{E})^2} .$$

The position-space diffusion coefficient is related to the field dependent diffusion coefficient in energy space by $D_s(E, \vec{r}, t) = \frac{D_F(E, \vec{r}, t)}{(e\vec{E})^2}$ [8].

$$S_{sb} \propto \frac{2\pi}{h} |F_k|^2 \left[ \frac{2F_k/\pi}{(\Omega_L - E_k - E^b_k - E_G)^2 + 4|F_k|^2} \right],$$

where

$$|F_k|^2 \approx \frac{e^2 E_{0k}^2 (x, y, z, t)}{m_0 \Omega_L} \left[ \frac{m_0 - m_e - 1}{2(E_G + 2\Delta_0)} \right].$$

$$\left(16\right)$$

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

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

Then the average energy gained over a collisionless flight (of order $\vec{E}^2$) is:

$$\langle E(t) - E_0 \rangle = (\vec{\partial}_F E)_{p_0} t + (\vec{\partial}_F^2 E)_{p_0} t^2 + \ldots$$

so $\langle E(t) - E_0 \rangle = e\vec{E} \cdot \vec{V}_p (\vec{p}_0) + (\vec{\partial}_F \vec{V}_p)_{p_0} \|\vec{p}_0\|$, and

$$V_F(E_0) = \left(\frac{dE}{dt}\right)_F = e\vec{E} \cdot \left[\vec{V}_g(\vec{p}_0) + (\vec{\partial}_F \vec{V}_g)_{p_0}\right] \|\vec{p}_0\|,$$

with $\vec{V}_d(E_0) = \int_{E_0} \vec{V}_g(\vec{p}_0) dS_p/\|\vec{V}_g\|$, where $dS_p/\|\vec{V}_g\|$ are the number of states in a surface element $dS_p$ in momentum space. From the above expressions the relation between the field dependent drift velocity and the drift velocity in coordinate space is obtained: $V_F(E_0) = e\vec{E} \cdot \vec{V}_d(E_0)$ and for the choice of laser field polarization

$$v_{ds}(E, \vec{r}, t) = \frac{V_F(E, \vec{r}, t)}{(e\vec{E})^2} .$$

The position-space diffusion coefficient is related to the field dependent diffusion coefficient in energy space by $D_s(E, \vec{r}, t) = \frac{D_F(E, \vec{r}, t)}{(e\vec{E})^2}$ [8].

$$\left(16\right)$$
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 $\frac{V(E,t)}{E^0}$ (eq.3) which has the physical meaning of energy dependent drift.

5. 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 the spatial dependence of the electromagnetic field is ignored and an electron-phonon system in a spatially uniform sinusoidal electric field of frequency $\Omega_1$ (dipole approximation) is considered. 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 conservation law for particle number and current density in energy space. Consequently the energy dependent current density is generalized to be energy and space dependent. The kinetic equation is generalized to include additional spatially dependent current densities on the left-hand side of the equation, thus yielding a multivariate Fokker-Planck type equation in the energy-position manifold.

From elementary arguments, without invoking expressions for the electron vector mean-free-path, related to momentum relaxation and mobility, the connection between the field dependent drift velocity in energy space and the drift velocity in real space is obtained.

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References
[1] Skupin S and Berge L 2006 Physica D 220 14
[2] Surdie L, Couarion A, Franco M, Lamouroux B, Prade B, Tzortzakis S and Mysyrowicz A 2002 Phys. Rev. Lett. 89 186601
[3] Apostolova T, Huang D, Alsing P M, McIver J and Cardimona D A 2002 Phys. Rev. B 66 075208
[4] Huang D, Apostolova T, Alsing P M and Cardimona D A 2004 Phys. Rev. B 69 75214
[5] Xu W 1998 Phys. Rev. B 57 12939
[6] Bringuier E 1997 Phys. Rev. B 56 5328
[7] Bringuier E 1998 Phys. Rev. B 57 2280
[8] Bringuier E 1995 Phys. Rev. B 52 8092
[9] Bringuier E 1996 Phys. Rev. B 54 8092
[10] Kurosawa T 1965 J. Phys. Soc. Jpn. 20 937