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Femtosecond laser irradiation induced-high electronic excitation in band gap materials: A quantum-kinetic model based on Boltzmann equation

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ABSTRACT

A quantum kinetic approach based on the Boltzmann equation is employed to describe the response of dielectric and semiconductor materials to high electronic excitation induced by laser irradiation. The formalism describes from the initial photo-ionization inter-band processes through free carrier absorption inducing additional impact ionization to the final heat up by electron–phonon coupling. Swift thermalization through electron–electron scattering, Auger recombination and formation of free excitons, their self-trapping and subsequent non-radiative decay are included. The energy exchange between the electrons and phonons are given by a separate equation for the lattice temperature where the rates of energy transfer from the electrons to the lattice per unit volume are defined quantum mechanically. As a result of our calculations the electron energy distribution function, average kinetic energy of the electron system and electron density are obtained as a function of laser intensity, laser photon energy (wavelength) and laser pulse duration. Examples of application in fs-laser irradiated-silica are discussed.

1. Introduction

Radiation-matter Physics has experienced a renewed interest in the last two decades. The reason stems from the current accessibility to radiation sources that can induce high electronic excitation in materials. These sources include intense electron and X-ray beams, swift heavy ion accelerators and remarkably intense fs laser facilities. Lasers were able to induce extremely high electronic densities only a couple of decades ago. Ultra-short (picosecond to femtosecond) laser pulses deliver very high peak power and are now accessible in several facilities worldwide. Compared to continuous lasers, ultra-short pulsed laser systems generate in the target high electronic density levels inducing much faster energy localization than energy diffusion. This turns them into a tool for controlled energy deposition that enables materials modification in a highly local and precise manner [1,2].

In this paper, we present our development of a consistent methodology to describe ultra-intense laser matter interaction applicable to bandgap materials that will encompass the transient dynamics of the processes occurring on different time scales. The main goal consists of identifying the dominant processes to simplify the formalism and obtain useful results. The final outcome is a feasible methodology to obtain results in acceptable computational times. The necessity of developing such a consistent methodology on the microscopic level arises from the fact that the optical and transport properties of semiconductors determine their applicability to devices such as photodetectors, transistors, and light emitters. The optical response of interband photodetectors decreases greatly when they are exposed to an intense laser field due to the incoherent creation of a large number of free electrons in the conduction band (CB). In addition, understanding the underlying microscopic paths of laser damage to semiconductors and dielectrics allows us to devise potential damage prevention or mitigation strategies.

2. Processes during and after a laser pulse

Irradiation by fs laser pulses leads to a myriad of processes in the material, mutually interconnected and relevant to the electronic evolution at different times. Next, we summarize these processes for fs laser irradiation of bandgap (non-metallic) materials:

- (Single- or multi-) photon ionization (PI) process, i.e., photon absorption.
- Quantum correction to the electron–phonon coupling referred to as phonon-assisted photon absorption or free-carrier absorption (FCA) from the laser field by the electrons already excited in the conduction band.

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- If a significant fraction of electrons get kinetic energies larger than the bandgap energy, they can excite an electron from the valence band to the conduction band, i.e., impact ionization (II) process.
- Excited electrons swiftly thermalize through electron–electron (EE) scattering process.
- Electron-impurity (EI) and electron–phonon (EP) scattering processes transfer energy from the electron system to the lattice of the material.
- Once the electron system is close to local equilibrium, exciton formation (XF) is possible and often dominant.
- Recombination processes such as Auger recombination (AR) and non-radiative exciton decay (NRXD) when enabled, are responsible for the reduction in the number of excited electrons and may induce energy transfer to the lattice.
- Typically, radiative recombination (RR), including direct intra-band recombination or radiative exciton decay, takes place over much longer timescales.
- Through EP, AR and NRXD, energy is transferred from the electron system to the lattice atoms. At this stage, thermal effects occur, such as ablation, heat diffusion and cooling, that in turn, give rise to permanent modifications (often very severe) in the irradiated material.

3. Boltzmann scattering equation method

The Boltzmann-type equation (BE) approach is used to calculate the temporal evolution of a given electron (hole) distribution corresponding to an energy in excess with respect to the bottom of the conduction band. This semi-classical approach implies that the evolution process is Markovian, i.e., the evolution of the system and the mean time-independent collision rate is calculated with the Fermi golden rule, which implies strict energy conservation. The effects of phase-space filling and band-gap renormalization should be included into the dynamic BE. The gap reduction can be determined by calculating the quasi-particle self-energy in a screened interaction perturbation treatment. The development of an improved quantum kinetic theory based on a generalized BE for the microscopic description of damage mechanisms induced by laser radiation must include the features of sub-picosecond electronic and structural events described in the previous section.

For the purposes described above the BE for electron distribution function used previously [3] is modified to include terms of time-dependent optical generation rate given in its semi-classical form as, e.g., in Ref. [4]. In the case of wide band gap dielectrics we include PI directly into the BE [5–7] as well as XF, II and AR. The connection between the electron occupation number, n_k^e, and the electron energy distribution function, f_k^e, is given in Ref. [8], $f_k^e = \rho_k n_k^e$, where $\rho_k^e = C_0\sqrt{E_k}$ is the reciprocal density of states with $C_0 = (2m_e^*)^{3/2}(2\pi^2\hbar^3)^{-1}$, being m_e^* the effective electron mass in the conduction zone. Then, we write the BE as,

$$\frac{\partial n_k^e}{\partial t} = W_k^{(\mathrm{in})(\alpha)}\left(1-n_k^e\right) - W_k^{(\mathrm{out})(\alpha)} n_k^e + G_{|\vec{k}|}^{\mathrm{PI}}\left(1-n_k^e\right) - W_k^{(\mathrm{exc})} n_k^e, \text{ with } W_k^{(\mathrm{exc})} = \frac{\sigma_{el}(E)}{\tau_f}, \tag{1}$$

where $W_k^{(\mathrm{in})(\alpha)}$ denotes the electron inter- and intra-band scattering with other electrons and with phonons and $W_k^{(\mathrm{exc})}$ denotes the exciton formation (further description in Section 3.2). Only the terms that are included additionally into the formalism described in [3] will be described next.

3.1. Photo-ionization (PI) and impact ionization (II)

The impact ionization scattering rates (and the inverse process of Auger recombination) [4,7,9–11] are given by:

$$W_k^{(\mathrm{in})(\mathrm{imp})} = \frac{2\pi}{\hbar}\sum_{\vec{k}',\vec{q}}\left|V^{(\mathrm{imp-in})}(q)\right|^2\left(1-n_{\vec{k}'}\right)n_{\vec{k}-\vec{q}}n_{\vec{k}'+\vec{q}}\delta \times\left(E_{\vec{k}}+E_{\vec{k}'}-E_{\vec{k}-\vec{q}}-E_{\vec{k}'+\vec{q}}\right), \tag{2}$$

$$W_k^{(\mathrm{out})(\mathrm{imp})} = \frac{2\pi}{\hbar}\sum_{\vec{k}',\vec{q}}\left|V^{(\mathrm{imp-out})}(q)\right|^2 n_{\vec{k}'}\left(1-n_{\vec{k}-\vec{q}}\right)\left(1-n_{\vec{k}'+\vec{q}}\right)\delta \times\left(E_{\vec{k}-\vec{q}}+E_{\vec{k}'+\vec{q}}-E_{\vec{k}}-E_{\vec{k}'}\right). \tag{3}$$

The static Thomas–Fermi screening effect is considered [1].

$$Q_s^2 = \frac{e^2}{\varepsilon_0\varepsilon_r}\frac{m^*}{\pi^2\hbar^2}\left(3\pi^2 n_{3D}\right)^{1/3}, \tag{4}$$

where ε_r is the average dielectric constant of the material, and n_{3D} is the concentration of conduction electrons in the bulk material. The Coulomb scattering potential is given by:

$$V^{(\mathrm{imp-in})}(q) = \left(\frac{\pi\hbar}{\tilde{I}_0}\right)^{1/2}\left(\frac{qe^2}{\varepsilon_0\varepsilon_r(q^2+Q_s^2)V}\right)\left(\frac{1}{m_{CB}^*}+\frac{1}{m_{VB}^*}\right)^{1/2}, \tag{5}$$

with effective ionization potential $\tilde{I}_0$ given in [6,7]. The PI term [6,7] is given by:

$$G_{\vec{|k|}}^{\mathrm{PI}} = \frac{2\pi}{\hbar}\sum_n\left|M_{\mathrm{pi},n}(\vec{k},\vec{k}')\right|^2\delta\left(E_{\vec{k}}^c-E_{\vec{k}}^h-N\hbar\Omega\right)n_{\vec{k}'}^h(1-n_{\vec{k}}) \approx W_{\mathrm{PI}}\pi\left(\frac{\hbar}{\sqrt{m^*}}\right)^3\sqrt{\frac{2}{E}}\frac{\Delta E}{\left(E-\frac{m_r}{m^*}(N\hbar\Omega-E_G)\right)^2+\Delta E^2}\times(1-n_k), \text{ since } n_{\vec{k}'}^h \approx 1 \tag{6}$$

$$\text{and } W_{\mathrm{PI}} = \sum_n\frac{1}{2\pi}\frac{\sqrt{2m_{cb}m_r^2}}{\hbar^3}\frac{V}{(2\pi)^3}\frac{2\pi}{\hbar}\left|M_{pi,n}(k)\right|^2\sqrt{E}. \tag{7}$$

where N is the number of photons involved in the multi-photon ionization process and the PI matrix element is given by (full expression and the rest of parameters are given in [7]),

$$\left|M_{pi,n}(k)\right|^2 = \frac{\hbar}{2\pi}\frac{2\pi^2\hbar^3 W_{pi,n}}{\sqrt{2m_{cb}m_r^2E_{pi,n}}}\frac{(2\pi)^3}{V}. \tag{8}$$

3.2. Exciton formation (XF) and non-radiative decay (NRXD)

The strong Coulomb interaction between an electron and a hole forces the pair to form an exciton. The additional collision integral for self-trapped exciton (STE)-formation that is added on the RHS of the BE (1) is given in Ref. [12]. The dimensionless cross section is an overlap integral of the STE-wavefunction, which is a Gaussian with a radius a_D, and the plane-wave conduction band electron function

$$\sigma_{el}(E) = \exp\left[-\frac{1}{2\pi}\frac{E}{E_{\mathrm{STE}}}\right], \quad \text{where } E_{\mathrm{STE}} = \frac{\hbar^2}{2m_c a_D^2} \tag{9}$$

In our approach, we use a coupled rate equation for exciton formation (decrement in the electron population in the conduction band) given by:

$$\left(\frac{dN_X}{dt}\right)_{XF} = -\frac{dN_C}{dt} = \int dE n_k W_k^{(\mathrm{exc})}, \tag{10}$$

where N_X represents the exciton density and N_C the electron density in the conduction band. We do not consider radiative exciton decay because the characteristic lifetimes are very long. However, excited excitons can decay non-radiatively [13] when enough lattice energy is available to overcome the kinetic barrier (ε) as depicted schematically in Fig. 1. We can account for the non-radiative decay simply