# Two-fluid plasma model for ultrashort laser-induced electron-hole nanoplasmas

B. Morel,* R. Giust, K. Ardaneh, and F. Courvoisier†
*FEMTO-ST institute, Univ. Bourgogne-Franche-Comté, CNRS,*
*15B avenue des Montboucons, 25030, Besançon Cedex, France*
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We present a two-fluid plasma model to describe localized interaction, at sub-micrometer scale, of highly focused ultra-short laser pulses within dielectrics and semiconductors. This model includes transport of the electron-hole plasma, electron-hole scattering, photoionization, impact ionization and interactions between charge carriers and phonons. We start with a plasma kinetic description and we consistently derive the two-fluid plasma model equations. Our numerical results highlight that transport of the electron-hole plasma can strongly affect the plasma density profile in the case of extremely localized interaction.

## I. INTRODUCTION

Ultrashort laser pulses are a powerful tool to induce modifications of materials[1–4]. Particularly in transparent dielectrics, ultrashort laser pulses can be used to locally modify, inside the material's bulk, the chemical structure, the index of refraction, the density of color centers, photopolymerize, generate nano-gratings, surface nanostructures or internal voids. A large number of application fields benefit from fundamental advances: surgery and biomedical applications, photonics, microfluidics, high-speed laser manufacturing[2,5–7].

Pushing forward these applications to nanometric structuring requires the support of numerical modelling[8]. Under laser-induced strong-field, bound electrons transit from the valence band to the conduction band[1,9,10], leaving a hole in the valence band. Particles of the electron-hole plasma are accelerated in the laser field, which results in the multiplication of the free carrier density via impact ionization, and potentially in the creation of a dense electron-hole plasma. Finally, at timescales much larger than several picoseconds, thermal and structural events takes place inside the material[1]. Our model is focused on the plasma density build-up, at timescales up to some picoseconds.

A large number of different models have been developed to study the propagation of ultra-short laser pulses (∼100 fs) in the high-intensity regime ($\sim 10^{14}$ W/cm$^2$) inside dielectrics and subsequent ionization. These models can be classified into two groups. In the first, several models were developed to study the filamentation regime inside transparent solids with propagation scales on the order of several tens of micrometers[11,12]. The pulse propagation model, such as the Unidirectional Pulse Propagation Equation[13], is derived from Maxwell's equations and coupled with a rate equation for free electron generation[14]. These models are well suited to describe the spatio-temporal evolution of the pulse under the different linear and nonlinear phenomena: Kerr and Raman effects, group velocity dispersion, diffraction, nonlinear ionization, plasma absorption and recombinations. They can be refined to describe non-paraxial propagation[15,16]. However, the derivation of the pulse propagation equation requires the assumptions of unidirectionality, local neutrality and transverse fields. These assumptions are not well suited to the case of nanoplasmas ($< 1\,\mu$m) induced by tightly focused beams[3].

In the second group, the models are intended to describe energy deposition in dielectrics at micrometric scale and are computationally very expensive because the full set of Maxwell equations is solved. Several descriptions of the plasma have been used: in references[17–19], plasma formation and heating is described by rate equations. A more refined model is the single fluid hydrodynamic approach[20,21]. An approach based on a two-temperature model (TTM) was used in Ref.[22].

In these previous works, the transport phenomena is conventionally neglected since valence holes are conventionally heavy compared to the effective electron mass ($m_{\mathrm{h}}^* = 5 - 10 m_{\mathrm{e}}$)[21,23,24]. Because of Coulomb attraction, transport of electrons and holes remain quasiidentical. The fluid of the heaviest particle (electron or hole) limit the transport of lightest one. If electron and hole have comparable light masses, then actual transport can be significant. Neglecting transport phenomena can be therefore inappropriate for materials with light holes such as sapphire[25], magnesium oxide[26], silicon[27], or zinc oxide[28]. Ultrafast transport at high-speed ($1.5\times10^6$ m/s) was recently observed experimentally in the latter material with pump-probe imaging-ellipsometry[28]. A density-dependent two temperature model (nTTM) was recently developed[29] and includes the electron-holes transport in semi-conductors. This model describes the evolution of free carrier density, carriers temperature, lattice temperature, density and energy transport, but hypothesizes plasma neutrality and same temperature for conduction electrons and valence holes.

Here, we propose a more general theoretical description of laser-induced excitation of dielectrics and semiconductors, that takes into account the transport phenomena and hence is valid for any effective mass ratio. We will particularly emphasize here the case where mass ratio is around 1, since it is the case that differs at most from the state of the art described above. In this case, we observed a significant transport, but also important consequences resulting from the holes dynamics (modification of the cut-off frequency, additional impact ionization, warm holes). Our fluid model includes 5-moments transport equations for each fluid, photoionization, impact ionization, electron-hole scattering and interactions between carriers and phonons. The approach is only valid when the band structure of the material remains, and is intended to describe the transient regime before the occurrence of nonthermal melting[30] or of the transformation of the plasma into warm dense matter[31–33].

The paper is organized as follows: in section II, we recall the two-fluid plasma model equations for an electron-hole plasma inside a bandgap material with parabolic energy bands, and we briefly review the different physical phenomena included in the model. In section III, we derive the source terms describing respectively electron-hole scattering, interactions between carriers and phonons, photoionization and impact ionization. Finally, in section IV, our numerical results of Bessel beam interaction with sapphire highlight that transport can significantly affect the plasma profile, throughout extremely localized interactions inducing nano-plasmas.

## II. THE TWO-FLUID PLASMA MODEL

The fluid model developed in this article is intended for materials that can be described by an isotropic parabolic band structure: the effective masses of conduction electrons and valence holes are independent of direction and position in Brillouin zone.

The fluid model can be derived consistently by calculating moments of the kinetic equation with respect to velocity[34] and by assuming a local thermodynamic equilibrium[35] for electrons and holes. This implies Maxwellian distributions for both particle species. The fluid description allows saving computational effort in comparison with the kinetic approach[36]. In our case, we consider that the plasma is sufficiently collisional to keep the particles distribution close to a Maxwellian distribution. The equations of two-fluid plasma model under balance laws form (*i.e.* a conservative form on the left side of fluid equations and a source term on the right one) are given by[37,38]:

$$\frac{\partial}{\partial t}\begin{bmatrix} n_\mathrm{e} \\ n_\mathrm{e}\mathbf{u}_\mathrm{e} \\ \epsilon_\mathrm{e} \\ n_\mathrm{h} \\ n_\mathrm{h}\mathbf{u}_\mathrm{h} \\ \epsilon_\mathrm{h} \end{bmatrix} + \nabla\cdot\begin{bmatrix} n_\mathrm{e}\mathbf{u}_\mathrm{e} \\ n_\mathrm{e}\mathbf{u}_\mathrm{e}\otimes\mathbf{u}_\mathrm{e} + \frac{p_\mathrm{e}}{m_\mathrm{e}^*}\mathbf{I} \\ (\epsilon_\mathrm{e}+p_\mathrm{e})\mathbf{u}_\mathrm{e} \\ n_\mathrm{h}\mathbf{u}_\mathrm{h} \\ n_\mathrm{h}\mathbf{u}_\mathrm{h}\otimes\mathbf{u}_\mathrm{h} + \frac{p_\mathrm{h}}{m_\mathrm{h}^*}\mathbf{I} \\ (\epsilon_\mathrm{h}+p_\mathrm{h})\mathbf{u}_\mathrm{h} \end{bmatrix} = \begin{bmatrix} 0 \\ \frac{n_\mathrm{e}q_\mathrm{e}}{m_\mathrm{e}^*}(\mathbf{E}+\mathbf{u}_\mathrm{e}\times\mathbf{B}) \\ n_\mathrm{e}q_\mathrm{e}\mathbf{u}_\mathrm{e}\cdot\mathbf{E} \\ 0 \\ \frac{n_\mathrm{h}q_\mathrm{h}}{m_\mathrm{h}^*}(\mathbf{E}+\mathbf{u}_\mathrm{h}\times\mathbf{B}) \\ n_\mathrm{h}q_\mathrm{h}\mathbf{u}_\mathrm{h}\cdot\mathbf{E} \end{bmatrix} + \begin{bmatrix} \mathbf{S}_\mathrm{e}^\mathrm{Coll} \\ \mathbf{S}_\mathrm{h}^\mathrm{Coll} \end{bmatrix} \tag{1}$$

The system of equations (1) corresponds to three conservation's laws for each fluids (electrons and holes): conservation of particles number, momentum and energy. The term with the divergence operator is called the transport term whereas the terms on the right member is called the source terms. For a specie of particles $a$ ($a = e$ for conduction electrons and $a = h$ for valence holes), $q_\mathrm{a}$ is the electric charge, $m_\mathrm{a}^*$ the effective mass, $n_\mathrm{a}$ the density, $\mathbf{u}_\mathrm{a}$ the mean velocity, $p_\mathrm{a}$ the scalar pressure, $\epsilon_\mathrm{a}$ the fluid energy density, $\mathbf{E}$ the electric field and $\mathbf{B}$ the magnetic field. $\mathbf{I}$ is the identity matrix and $\otimes$ is tensor product. To obtain the system of equations (1), we additionally made the assumption that electrons and holes fluids are non-viscous. The effects of viscosity are usually unimportant in ultra-fast dynamics[39]. It allows us to consider scalar pressures instead of pressure tensors.

The system of equations for electrons and hole fluids is closed by assuming local thermodynamic equilibrium (Maxwellian distribution) for both species:

$$\epsilon_\mathrm{a} \equiv \frac{p_\mathrm{a}}{\gamma-1} + \frac{1}{2}n_\mathrm{a}m_\mathrm{a}^*{\mathbf{u}_\mathrm{a}}^2 \tag{2}$$

where $\gamma$ is the adiabatic index. The local thermodynamic equilibrium allows linking pressure and temperature through the ideal gas law[35]: $p_a = n_a k_B T_a$, where $T_a$ is the temperature and $k_B$ the Boltzmann constant.

The terms $\mathbf{S}_e^{Coll}$ and $\mathbf{S}_h^{Coll}$ are the collisional terms for electron and hole fluids. We use the same general method as in Ref.[40,41] in order to include different physical phenomena, *i.e.* we split the collisional terms of Eq. (1) in several terms:

$$\underbrace{\begin{bmatrix}\mathbf{S}_e^{Coll}\\ \mathbf{S}_h^{Coll}\end{bmatrix}}_{\equiv \mathbf{S}^{Coll}} \equiv \underbrace{\begin{bmatrix}\mathbf{S}_e^{e-h}\\ \mathbf{S}_h^{e-h}\end{bmatrix}}_{\equiv \mathbf{S}^{e-h}} + \underbrace{\begin{bmatrix}\mathbf{S}_e^{Imp-e}\\ \mathbf{S}_h^{Imp-e}\end{bmatrix}}_{\equiv \mathbf{S}^{Imp-e}} + \underbrace{\begin{bmatrix}\mathbf{S}_e^{Imp-h}\\ \mathbf{S}_h^{Imp-h}\end{bmatrix}}_{\equiv \mathbf{S}^{Imp-h}} + \underbrace{\begin{bmatrix}\mathbf{S}_e^{Ph}\\ \mathbf{S}_h^{Ph}\end{bmatrix}}_{\equiv \mathbf{S}^{Ph}} + \underbrace{\begin{bmatrix}\mathbf{S}_e^{e-p}\\ \mathbf{S}_h^{e-p}\end{bmatrix}}_{\equiv \mathbf{S}^{e-p}} + \underbrace{\begin{bmatrix}\mathbf{S}_e^{h-p}\\ \mathbf{S}_h^{h-p}\end{bmatrix}}_{\equiv \mathbf{S}^{h-p}} \tag{3}$$

where the superscript *e-h* is related to electrons-holes scattering, *e-p* to interactions between electrons and phonons, *h-p* to interactions between holes and phonons, *Ph* to photoionization, *Imp-e* to electron-initiated impact ionization and *Imp-h* to hole-initiated impact ionization.

The electric and magnetic fields in Eq. (1) are determined by Maxwell equations:

$$\nabla \cdot \varepsilon_r \mathbf{E} = \frac{1}{\varepsilon_0}\left[q_e n_e + q_h n_h\right] \tag{4}$$

$$\nabla \cdot \mathbf{B} = 0 \tag{5}$$

$$\nabla \times \mathbf{E} = -\frac{\partial \mathbf{B}}{\partial t} \tag{6}$$

$$\nabla \times \mathbf{B} = \mu_0 \mathbf{J} + \frac{1}{c^2}\frac{\partial \varepsilon_r \mathbf{E}}{\partial t} \tag{7}$$

where $\varepsilon_0$ and $\mu_0$ are respectively the vacuum permittivity and permeability. $c = (\varepsilon_0 \mu_0)^{-1/2}$ is the speed of light. $\varepsilon_r$ is the relative permittivity of the background dielectric. The total current density $\mathbf{J}$ is composed of the contribution from conduction electrons and of the one from valence holes:

$$\mathbf{J} = q_e n_e \mathbf{u}_e + q_h n_h \mathbf{u}_h \tag{8}$$

In our description, we consider an electron-hole plasma localized in background dielectric of relative permittivity $\varepsilon_r$. The effect of bound charges is also taken into account through relative permittivity $\varepsilon_r$ whereas the effect of charge carriers is included through the current density $\mathbf{J}$ of Eq. (8).

We finally remark that, here, the medium response to an electromagnetic excitation is taken into account through a fluid approach. This approach is valid only when the spatial scales are larger than the Debye length[42]. The typical densities involved in numerical results of section IV are around $10^{21}$ cm$^{-3}$ and the temperatures are in a range of 1-100 eV, leading to a Debye length between 0.2 nm and 2 nm.

## III. COLLISIONAL SOURCE TERMS

In this section, we specify the source terms $\mathbf{S}_e^{Coll}$ and $\mathbf{S}_h^{Coll}$ of Eq. (3). We include the different physical phenomena one by one in our model.

### A. Electron-hole scattering

We start with the elastic scattering between conduction electrons and valence holes. We have chosen to include this phenomenon via the Bhatnagar-Gross-Krook (BGK) approach[43]. This approach assumes that the elastic scattering restores a local equilibrium with an exponential decay in time. This description usually gives a first approximation to the problem under consideration[35] and is relatively easy to link to the Drude model of plasmas[44]. The inclusion of the BGK term in the two-fluid plasma equations is performed via the following source term[45]:

$$\mathbf{S}^{\mathbf{e-h}} = \begin{bmatrix}\mathbf{S}_e^{\mathbf{e-h}}\\ \mathbf{S}_h^{\mathbf{e-h}}\end{bmatrix} = \begin{bmatrix} 0 \\ -n_e \nu_{eh}\frac{\mu}{m_e^*}(\mathbf{u}_e - \mathbf{u}_h) \\ -\frac{\kappa \nu_{eh} n_e}{\gamma-1}(T_e - T_h) + \frac{\kappa \nu_{eh} n_e}{2}\left(m_e^* \mathbf{u}_e{}^2 - m_h^* \mathbf{u}_h{}^2 + (m_h^* - m_e^*)\mathbf{u}_e \cdot \mathbf{u}_h\right) \\ 0 \\ -n_h \nu_{he}\frac{\mu}{m_h^*}(\mathbf{u}_h - \mathbf{u}_e) \\ -\frac{\kappa \nu_{he} n_h}{\gamma-1}(T_h - T_e) + \frac{\kappa \nu_{he} n_h}{2}\left(m_h^* \mathbf{u}_h{}^2 - m_e^* \mathbf{u}_e{}^2 + (m_e^* - m_h^*)\mathbf{u}_h \cdot \mathbf{u}_e\right) \end{bmatrix} \tag{9}$$

where $\mu = \frac{m_e^* m_h^*}{m_e^* + m_h^*}$ is the reduced mass, $\kappa = \frac{2\mu}{m_e^* + m_h^*}$ and $\nu_{eh}$ is the scattering frequency for momentum transfer from electrons to holes whereas $\nu_{he}$ is from holes to electrons. Eq. (9) is symmetrical to the permutation $e \leftrightarrow h$, and conservation of momentum and energy imposes the following condition[45]:

$$n_{\mathrm{e}}\nu_{\mathrm{eh}} = n_{\mathrm{h}}\nu_{\mathrm{he}} \tag{10}$$

### B. Interactions between charge carriers and phonons

As in Ref.[41], we consider two scattering phenomena for the interactions between charge carriers and phonons. The first one is the charge carrier - phonon elastic scattering, where charge carriers undergo a momentum change due to phonon emission/absorption. The second phenomenon is the carrier-phonon-photon interaction. In these two phenomena, the loss of energy to the phonon bath is negligible at sub-picosecond timescale[9,33]. The impact of these scattering events on the conduction electron fluid and on the valence hole fluid is a modification of the distribution of the particle, *i.e.,* a reduction of the mean velocity and an increase of the thermal energy. We include these phenomena in a classical way (*i.e.,* in a non-quantum treatment) via frictions terms[23]:

$$\mathbf{S}^{\mathbf{e-p}} = \begin{bmatrix} \mathbf{S}_{\mathrm{e}}^{\mathbf{e-p}} \\ \mathbf{S}_{\mathrm{h}}^{\mathbf{e-p}} \end{bmatrix} \equiv \begin{bmatrix} 0 \\ -n_{\mathrm{e}}(\nu_{\mathrm{epp}} + \nu_{\mathrm{ep}})\mathbf{u}_{\mathrm{e}} \\ 0 \\ 0 \\ \mathbf{0} \\ 0 \end{bmatrix} \tag{11}$$

where $\nu_{\mathrm{epp}}$ is the electron-phonon-photon interaction rate and $\nu_{\mathrm{ep}}$ is the electron-phonon scattering rate. Similarly, we have for the hole fluid:

$$\mathbf{S}^{\mathbf{h-p}} = \begin{bmatrix} \mathbf{S}_{\mathrm{e}}^{\mathbf{h-p}} \\ \mathbf{S}_{\mathrm{h}}^{\mathbf{h-p}} \end{bmatrix} \equiv \begin{bmatrix} 0 \\ \mathbf{0} \\ 0 \\ 0 \\ -n_{\mathrm{h}}(\nu_{\mathrm{hpp}} + \nu_{\mathrm{hp}})\mathbf{u}_{\mathrm{h}} \\ 0 \end{bmatrix} \tag{12}$$

where $\nu_{\mathrm{hpp}}$ is the hole-phonon-photon interaction rate and $\nu_{\mathrm{hp}}$ is the hole-phonon scattering rate.

In this description with friction terms, the total energies $\epsilon_{\mathrm{a}}$ of the fluids do not change (no term in components 3 and 6 of Eq. (11) and Eq. (12)). The scattering induces a conversion of directed energy $\frac{1}{2}n_{\mathrm{a}}m_{\mathrm{a}}^{*}\mathbf{u}_{\mathrm{a}}{}^{2}$ into thermal energy $\frac{p_{\mathrm{a}}}{\gamma-1}$. It also induces a weakening of the current density $\mathbf{J}$, thus absorbing energy from the electromagnetic fields.

We note that here, we discard the energy transfer from the carriers to the phonon bath since it is effective at timescales of several picoseconds while our model is intended to model typically sub-picosecond dynamics[9,33].

### C. Electron-electron and hole-hole scattering

Electron-electron and hole-hole elastic scattering do not affect fluid equations (1)[29,35]. The reason is that in the fluid approach, the shape of the distribution is fixed to a Maxwellian one, before and after the scattering process. In addition, during a scattering event, the change of momentum (resp. energy) of a particle is equal to the change of momentum (resp. energy) of the other particle. This implies that the total momentum (resp. energy) of the fluid remain constant during elastic scattering between particles of same specie[35].

### D. Photoionization

Photoionization, impact ionization and recombination can be considered as inelastic collisions.[35]. However, it is conventionally difficult to rigorously describe inelastic collisions in the framework of the fluid description[46]. In this section, we insert photoionization (promotion of electrons in the conduction band by absorption of field energy) in the two-fluid plasma model. Impact ionization will be modelled in the following section. We start by defining $n_{\mathrm{ev}}$ as the density of electron available in the valence band: this number is a function of time and space. We set $n_0$ as the initial density of electron available in the valence band, and $\alpha^{\mathrm{Ph}}$ as the photoionization rate in $\mathrm{m}^{-3}\mathrm{s}^{-1}$.

To include photoionization, we make the following assumptions: (i) the number of created fluid particles (electrons and holes) through photoionization is given by rate equation used in Ref.[17] ($\frac{\partial}{\partial t}n_{\mathrm{e}} = \frac{n_{\mathrm{ev}}}{n_0}\alpha^{\mathrm{Ph}}$). We use the Keldysh ionization rate in solid[47] (appendix A), but it could be replaced by more sophisticated models[48] without changing the overall architecture of our model. (ii) as in Ref.[49], we consider that during photoionization, valence electrons are promoted to the bottom of the conduction band. This is equivalent to neglecting the residual energy transfer to charge carriers after photoionization: fluid particles are generated with zero energy ($p_{\mathrm{a}} = 0$ and $\mathbf{u}_{\mathrm{a}} = \mathbf{0}$).

The source term for photoionization of Eq. (3) is therefore given by:

$$\mathbf{S}^{\mathbf{Ph}} \equiv \begin{bmatrix} \mathbf{S}_{\mathrm{e}}^{\mathbf{Ph}} \\ \mathbf{S}_{\mathrm{h}}^{\mathbf{Ph}} \end{bmatrix} \equiv \begin{bmatrix} \frac{n_{\mathrm{ev}}}{n_0}\alpha^{\mathrm{Ph}} \\ \mathbf{0} \\ 0 \\ \frac{n_{\mathrm{ev}}}{n_0}\alpha^{\mathrm{Ph}} \\ \mathbf{0} \\ 0 \end{bmatrix} \tag{13}$$

For each electron-hole pair generated, one valence electron must be removed. This leads to the following rate equation for the density of valence electrons:

$$\frac{\partial}{\partial t}n_{\mathrm{ev}} = -\frac{n_{\mathrm{ev}}}{n_0}\alpha^{\mathrm{Ph}} \tag{14}$$