# Unified Model for the Free-Electron Avalanche in Laser-Irradiated Dielectrics

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(Received 30 October 2003; published 6 May 2004; publisher error corrected 11 May 2004)

We develop a model describing the free-electron generation in transparent solids under high-intensity laser irradiation. The multiple rate equation model unifies key points of detailed kinetic approaches and simple rate equations to a widely applicable description, valid on a broad range of time scales. It follows the nonstationary energy distribution of electrons on ultrashort time scales as well as the transition to the asymptotic avalanche regime for longer irradiations. The role of photoionization and impact ionization is clarified in dependence on laser pulse duration and intensity.

DOI: 10.1103/PhysRevLett.92.187401 PACS numbers: 78.47.+p, 02.60.Lj, 77.90.+k

We address the problem of high-intensity laser interaction with transparent solids, a problem that is most relevant to materials processing applications using ultrashort laser pulses. Despite the plethora of experimental and theoretical papers which have appeared on the subject, significant controversies remain [1–6]. Part of the problem is related to the fact that the complicated cascade of physical processes that follow intense laser irradiation has either been modeled in impractically complicated detail or, in more transparent approaches, largely oversimplified. Here, we seek to bring unity to the field by developing a widely applicable model which includes key points of detailed kinetic approaches in a most practical description, valid on a broad range of time scales. We introduce the multiple rate equation (MRE) consisting of a set of coupled ordinary differential equations. It represents the first description of the transient free-electron density which keeps track of the electrons energy distribution while maintaining the conceptual and analytic simplicity of standard rate equations. The solution shows the transient nonstationary energy distribution of electrons on ultrashort time scales and follows the transition to the asymptotic avalanche behavior. The model clarifies the dependence of the role of different ionization processes on laser pulse duration and intensity.

The transient free-electron density is a fundamental parameter in numerous experimental and theoretical investigations. Usually, it is described by a simple rate equation, combining the rate of photoionization $\dot{n}_{\mathrm{pi}}$ with the rate of impact ionization, assumed to depend on the total free-electron density $n_{\mathrm{total}}$:

$$\frac{dn_{\mathrm{total}}}{dt} = \dot{n}_{\mathrm{pi}}(E_L) + \alpha(E_L)\, n_{\mathrm{total}}\,. \tag{1}$$

Because of photoionization, depending directly on the amplitude of the electric laser field $E_L$, electrons are shifted from the valence band into the conduction band [7]. In contrast, electron–electron impact ionization is caused by a free electron already existing in the conduction band. If its kinetic energy is sufficiently large, it may transfer part of it to an electron in the valence band, such that the latter is enabled to overcome the ionization potential [8,9]. The avalanche coefficient $\alpha$ depends on the effective energy gain of the free electron in the electric laser field $E_L$ and can be estimated by approximative models or is taken as a fit parameter. Equation (1) was proposed and verified for laser pulses in the nanosecond regime (see, for instance, Refs. [1,10], and references therein).

While experimental studies applying Eq. (1) have lead to contradictory results, refuting [1,2] or emphasizing [3–5] the importance of the electron avalanche in the picosecond regime and below, fundamental doubts exist whether this standard rate Eq. (1) is applicable in general on ultrashort time scales [6,11,12]. One basic assumption of Eq. (1) is that impact ionization depends directly on the total density of the free electrons. However, also the *energy* of a particular electron plays an important role: Photoionization generates electrons with *low* kinetic energy in the conduction band while impact ionization requires electrons of *high* kinetic energy. This additional energy is absorbed from the laser light by intraband absorption. If this absorption process takes time comparable to the laser pulse duration, it is obvious that Eq. (1) is oversimplified. At least until the shape of the transient distribution function of the electrons in the conduction band becomes stationary, the energy distribution of the electrons is crucial for the probability of impact ionization. However, its explicit calculation using full kinetic approaches is not practical for numerous cases where only the transient energy-averaged total electron density is of interest. The problem is enhanced by contradictory conclusions about the applicability of Eq. (1) resulting from different interpretations of the involved results of complex kinetic approaches [5,6].

The MRE model, introduced in the following, brings light into this discussion, providing a direct possibility for estimating the role of the impact ionization avalanche and the validity of Eq. (1) in dependence on laser pulse duration and intensity. It considers essential features of the transient electron energy distribution describing the resulting delay of the electron avalanche in a most basic way, formally considering discrete energy levels but neglecting relaxation processes such as electron–electron collisions and electron–phonon collisions [13]. Figure 1 outlines the axis of kinetic energy $\varepsilon$ in the conduction band (CB) of a dielectric, together with the processes involved in the generation of free electrons marked by arrows at the corresponding energies. Basically, the cycle of ionization is as follows: Photoionization generates electrons with a certain ionization rate $\dot{n}_{\rm pi}$ at the lower edge of the conduction band, i.e., with energy $\varepsilon_0 \approx 0$. An electron at energy $\varepsilon_0$ may absorb a single photon from the laser light with probability $W_{\rm 1pt}(\varepsilon_0)$. The resulting kinetic energy of the electron reads $\varepsilon_1 := \varepsilon_0 + \hbar\omega_L$, where $\omega_L$ is the laser frequency. In the same manner, further discrete energy levels $\varepsilon_{j+1} := \varepsilon_j + \hbar\omega_L$ are defined. When $k = \lceil \varepsilon_{\rm crit}/\hbar\omega_L \rceil$ photons have been absorbed, where $\lceil x \rceil$ denotes the integer above $x$, the electrons energy $\varepsilon_k = \varepsilon_0 + k\hbar\omega_L$ exceeds the critical energy for impact ionization $\varepsilon_{\rm crit}$. For electrons with $\varepsilon > \varepsilon_{\rm crit}$ impact ionization occurs with a probability $\tilde{\alpha}$. Through this process the kinetic energy will be reduced and a second electron is shifted from the valence band (VB) into the conduction band. Both electrons will then have a small kinetic energy, which can be assumed to be comparable to $\varepsilon_0$.

Defining the density $n_j$ as the density of electrons at energy $\varepsilon_j$, one can describe this process with the following system of rate equations, representing the multiple rate equation (MRE):

$$\begin{aligned} \dot{n}_0 &= \dot{n}_{\rm pi} + 2\,\tilde{\alpha}\,n_k - W_{1pt}(\varepsilon_0)\,n_0, \\ \dot{n}_1 &= W_{\rm 1pt}(\varepsilon_0)\,n_0 - W_{\rm 1pt}(\varepsilon_1)\,n_1, \\ &\vdots \\ \dot{n}_{k-1} &= W_{\rm 1pt}(\varepsilon_{k-2})\,n_{k-2} - W_{\rm 1pt}(\varepsilon_{k-1})\,n_{k-1}, \\ \dot{n}_k &= W_{\rm 1pt}(\varepsilon_{k-1})\,n_{k-1} - \tilde{\alpha}\,n_k, \qquad \text{with } k = \left\lceil \frac{\varepsilon_{\rm crit}}{\hbar\omega_L} \right\rceil. \end{aligned} \tag{2}$$

Adding up these equations yields

$$\frac{dn_{\rm total}}{dt} = \dot{n}_{\rm pi}(E_L) + \tilde{\alpha}\,n_k, \tag{3}$$

with $n_{\rm total} = \sum_{j=1}^{k} n_j$. The difference in the last term of Eq. (3) compared to Eq. (1) is substantial: while in Eq. (1) the impact ionization is assumed to depend on the *total* free-electron density $n_{\rm total}$, Eq. (3) considers the fact that only those electrons which bear sufficiently high energy may produce impact ionization. Note that the density of high-energy electrons $n_k$ formally includes also electrons with energy $\varepsilon > \varepsilon_k$. In case of a *stationary* free-electron distribution function, the fraction of high-energy electrons $n_k/n_{\rm total}$ is *temporally constant* and Eq. (3) reduces to Eq. (1) with $\alpha = \tilde{\alpha}\,n_k/n_{\rm total}$. Thus, equation system (2) includes the standard rate Eq. (1) as a special case.

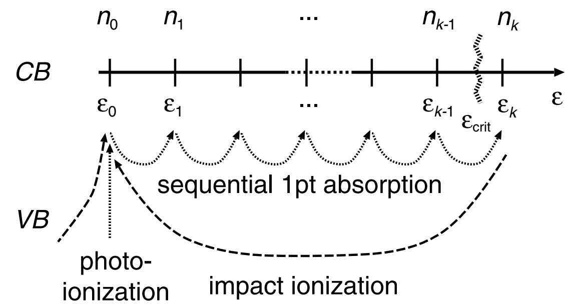


FIG. 1. Illustration of the processes providing changes in the density and the energy, respectively, of free electrons in the conduction band of a dielectric.

The MRE (2) may be solved with the help of Laplace transform. The initial densities $n_j$ can be neglected. In the following example, constant one-photon-absorption probabilities $W_{\rm 1pt}(\varepsilon_j) = W_{\rm 1pt}$ for $j = 0 \ldots k$ are assumed. The $\mathcal{L}$-transformed function $\eta_k(s)$ of $n_k(t)$ reads:

$$\eta_k(s) = \frac{W_{\rm 1pt}^k}{(s + \tilde{\alpha})(s + W_{\rm 1pt})^k - 2\tilde{\alpha}W_{\rm 1pt}^k} \cdot \frac{\dot{n}_{\rm pi}}{s}. \tag{4}$$

Its inverse transform can be performed by the sum of residua of $\eta_k(s)\exp(st)$. The solution can be found analytically for the case that $\tilde{\alpha} \gg W_{\rm 1pt}$, which is a similar but weaker assumption than the often applied so-called "flux-doubling" model [5,14]. In this case the $k+2$ poles of $\eta_k(s)$ are $s_0 = 0$, $s_\alpha \simeq -\tilde{\alpha}$, and $k$ complex poles $s_{1..k} \simeq (\sqrt[k]{2} - 1)\,W_{\rm 1pt}$.

For brevity only the asymptotic solution for comparably long times is shown here. This is determined by the largest real positive pole of Eq. (4), $s_1 \simeq (|\sqrt[k]{2}| - 1)\,W_{\rm 1pt}$. For times $t \gg s_1^{-1}$ the inverse transform yields the density of high-energy electrons in the asymptotic regime,

$$n_k(t) = \frac{\dot{n}_{\rm pi}}{2k(1 - \sqrt[k]{1/2})\tilde{\alpha}} \exp[(|\sqrt[k]{2}| - 1)\,W_{\rm 1pt}\,t] + \ldots. \tag{5}$$

After inclusion in Eq. (3) and integration, the asymptotic total free-electron density is obtained as a function of time:

$$n_{\rm total}(t) = \frac{\dot{n}_{\rm pi}/W_{\rm 1pt}}{2k(\sqrt[k]{2} - 2 + \sqrt[k]{1/2})} \times \exp[(|\sqrt[k]{2}| - 1)\,W_{\rm 1pt}\,t] \qquad \text{for } t \gg t_{\rm MRE} := [(|\sqrt[k]{2}| - 1)\,W_{\rm 1pt}]^{-1}. \tag{6}$$

Thus, for long times $t \gg t_{\rm MRE}$, the total electron density grows exponentially, as predicted by the simple avalanche model. Comparison with (1) yields the corresponding avalanche coefficient

$$\alpha_{\rm asymp} = (|\sqrt[k]{2}| - 1)\,W_{\rm 1pt}, \tag{7}$$

which describes the growth of the total electron density for times much larger than $t_{\rm MRE} = \alpha_{\rm asymp}^{-1}$.

For times in the range of $t_{\rm MRE}$ and below, i.e., *before* the asymptotic regime (6) is reached, the complete inverse transform of Eq. (4) should be performed. Another possibility to figure out the behavior of electrons on ultrashort time scales is the numerical solution of the MRE (2). For the following examples we choose parameters similar to those applied in Ref. [6] in order to allow direct comparison with the kinetic approach. The critical energy for impact ionization is given by [6]

$$\varepsilon_{\mathrm{crit}} = (1 + \mu/m_{\mathrm{VB}})(E_{\mathrm{gap}} + \langle\varepsilon_{\mathrm{osc}}\rangle). \tag{8}$$

Here, the first factor accounts for the conservation of electron momentum [8]; the present form follows for parabolic valence and conduction bands. $\mu$ is the reduced mass of the effective electron mass in the valence band, $m_{\mathrm{VB}}$, and in the conduction band, $m_{\mathrm{CB}}$. The second factor in Eq. (8) is the effective ionization potential given by the bandgap-energy $E_{\mathrm{gap}}$ enhanced by the mean oscillation energy $\langle\varepsilon_{\mathrm{osc}}\rangle = e^2E_L^2/(4\mu\omega_L^2)$ [7]. We assume the effective masses $m_{\mathrm{VB}}$ and $m_{\mathrm{CB}}$ both equal to the free-electron mass and a bandgap of $E_{\mathrm{gap}} = 9$ eV. The applied laser is chosen with a wavelength $\lambda = 500$ nm, corresponding to a photon energy of $\hbar\omega_L = 2.48$ eV. Assuming electric laser field amplitudes up to $E_L = 1 \times 10^{10}$ V/m, the critical energy of impact ionization according to Eq. (8) lies between 13.5 and 14.5 eV, hence, the MRE (2) consists of $k + 1 = 7$ equations. The rate of photoionization $\dot{n}_{\mathrm{pi}}$ is taken from Ref. [7] and corresponds to the case of multiphotonionization for the applied field strengths. The probability of impact ionization $\tilde{\alpha}$ can be estimated from the corresponding collision term given in Ref. [6] and lies in the range of $\tilde{\alpha} \approx 1 \times 10^{15}$ s$^{-1}$. As long as $\tilde{\alpha} \gg W_{\mathrm{1pt}}$, the exact value of $\tilde{\alpha}$ plays no role for the total free-electron density, as Eq. (6) shows for the asymptotic case. $W_{\mathrm{1pt}}(\varepsilon_j)$ is chosen to be $W_{\mathrm{1pt}} = 3.5 \times 10^{-7}E_L^2$ m$^2$/V$^2$ s, independent of the electron energy $\varepsilon$. This expression compares well with the mean value of the one-photon absorption probability for $SiO_2$ in Refs. [6,15].

Figure 2 shows the transient fraction of high-energy electrons $n_k/n_{\mathrm{total}}$ multiplied with $\tilde{\alpha}$ (therewith being independent on the choice of $\tilde{\alpha}$). It reflects the temporal evolution of the shape of the free-electron distribution function for ultrashort times and its development towards the asymptotic stationary long-time behavior. Depending on electric laser field, the time to reach the stationary regime and thus a constant fraction of high-energy electrons is in the range of several hundreds of femtoseconds. Below this time scale the fraction of high-energy electrons is much lower than its asymptotic value. Figure 2 shows also the normalized fraction of electrons with energy above $\varepsilon_{\mathrm{crit}}$ resulting from the full kinetic calculation of Kaiser *et al.* [6] for an electric field of $E_L = 50$ MV/cm. The transient nonstationary electron distribution is very well imitated by the MRE model (2). For times much larger than $t_{\mathrm{MRE}}$, when the stationary regime is reached and Eq. (1) can be assumed to be valid, the fraction $\tilde{\alpha}n_k/n_{\mathrm{total}}$ provides the avalanche coefficient for Eq. (1); compare Eq. (3). The dotted lines in Fig. 2 correspond to the analytically calculated asymptotic value of $\alpha(E_L)$ according to Eq. (7), assumed to be valid after $t \gg t_{\mathrm{MRE}}$. The MRE model (2) thus provides a comparably simple possibility both to consider the nonstationary electron distribution in dielectrics during ultrashort laser irradiation and to follow the transition to the asymptotic avalanche regime for longer times. The transition time $t_{\mathrm{MRE}}$ is much larger than the results of Ref. [5] suggest, which were obtained *excluding* photoionization and assuming some initial shape of the electron distribution. Under such conditions also the MRE provides an immediately established avalanche. However, in most practical cases photoionization cannot be simply excluded and its role is essential for the nonstationarity of the electron distribution on ultrashort time scales.

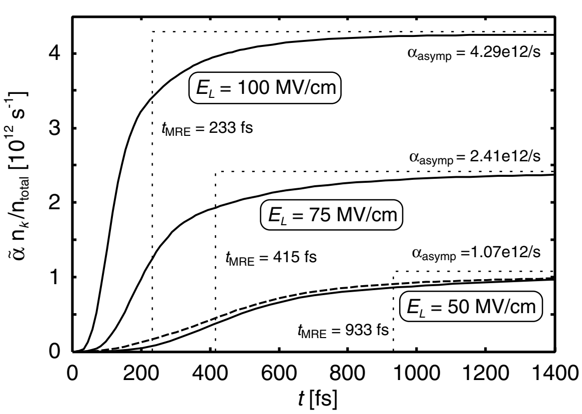


FIG. 2. Temporal evolution of the fraction of high-energy electrons $\tilde{\alpha}n_k/n_{\mathrm{ges}}$ for different laser field amplitudes calculated by the MRE (2) (solid lines). The asymptotical values coincide with the avalanche coefficient $\alpha_{\mathrm{asymp}}$ from Eq. (7). For times up to the range of $t_{\mathrm{MRE}}$ the fraction of high-energy electrons strongly changes in time. The dashed line shows the normalized fraction of high-energy electrons calculated with the full kinetic approach from Ref. [6] for $E_L = 50$ MV/cm.

The MRE is widely applicable to a broad variety of experimental and theoretical research, ranging from investigations of laser induced dielectric breakdown to fundamental research of laser-matter interaction. For example, in Ref. [2] single-shot time-resolved experiments were performed to study the free-electron density evolution in dielectrics. These experiments could be successfully interpreted only when neglecting the contribution of the electron avalanche, i.e., the second term in Eq. (1). The MRE explains the apparent lack of the free-electron avalanche by the small impact ionization rate due to the nonstationary electron distribution. For pulse durations below $t_{\mathrm{MRE}}$ the low fraction of high-energy electrons leads to a low contribution of the electron impact ionization as shown in Fig. 3. Here, the transient total free-electron density $n_{\mathrm{total}}(t)$ was calculated with different models for the case of a Gaussian-shaped laser pulse of 300 fs full-width-half-maximum (FWHM) duration and a maximum electric laser field of $E_{L,0} = 70$ MV/cm. The MRE reproduces the small contribution of the impact ionization resulting from the full kinetic approach very well. In contrast, the standard rate equation strongly overestimates this contribution. The experiments with picosecond pulses in Ref. [2] were performed at lower intensities so that as well $t_{\mathrm{MRE}}$ attains

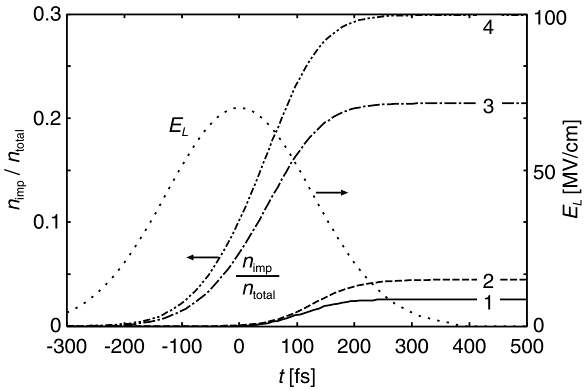


FIG. 3. Contribution of impact ionization to the total free electron density generated by a 300 fs FWHM Gaussian laser pulse with a maximum electric field of $E_{L,0} = 70$ MV/cm. Curve 1 was calculated with the MRE (2), curve 2 is the result of the full kinetic approach from Ref. [6], curve 3 was calculated with the standard rate Eq. (1) using $\alpha(E_L) = \alpha_{asymp}$ according to Eq. (7), and curve 4 is the result of Eq. (1) with a common estimation for the avalanche parameter, $\alpha_{est} = W_{1pt}\,\hbar\omega_L/\varepsilon_{crit}$.

the picosecond regime, leading again to a low impact ionization rate. In the same manner the MRE explains earlier results of Jones *et al.* [1] refuting the avalanche model (1) at visible wavelength and picosecond pulses around breakdown threshold.

In the case of high intensities, $t_{MRE}$ becomes small and lies in the femtosecond regime. Then, a significant contribution of the impact ionization avalanche can be obtained also for subpicosecond laser pulses. For very high intensities, when $\langle\varepsilon_{osc}\rangle$ may become larger than the photon energy, the avalanche may also be enhanced by intraband absorption of higher order. In theoretical investigations, for instance in studies of nonlinear propagation of ultrashort high-intensity laser pulses in transparent media [16,17], often an avalanche parameter $\alpha_{est,gap} = W_{1pt}\,\hbar\omega_L/E_{gap}$ is applied, which takes intuitively into account the probability of energy absorption by free electrons up to the energy necessary to perform impact ionization. Applying $\varepsilon_{crit}$ instead of $E_{gap}$, $\alpha_{est}$ is slightly larger than $W_{1pt}/k$, which compares with the limit for $k \to \infty$ of $\alpha_{asymp} \to \ln(2)\,W_{1pt}/k$. The factor ln(2) takes into account that the number of electrons is doubled in each impact ionization event. Thus, the MRE provides a correction to the usual estimation; it is able to *calculate* the avalanche parameter valid in the asymptotic long-time regime from a physically motivated model.

In summary, we developed a widely applicable model which describes the free-electron density evolution in the conduction band of a dielectric under ultrashort laser irradiation. The multiple rate Eq. (2), consisting of an ordinary differential equation system of rate equations, provides a unique tool for numerous theoretical and experimental investigations where the transient free-electron density is an important parameter. It gives insight into the short-time physics in dielectrics keeping track of the energy distribution of the free electrons as up to now only realized by kinetic approaches. Its asymptotic solution (6) provides the avalanche parameter $\alpha$, entering the standard rate equation, and shows directly the condition $t \gg t_{MRE}$ under which Eq. (1) is applicable. Moreover, the MRE describes also the *transition* between these physically different regimes of nonstationarity on ultrashort time scales and the asymptotic avalanche behavior on longer time scales.

Stimulating discussions with A. Kaiser and J. Krug and valuable suggestions of A. Cavalleri, P. Mulser, S. Anisimov, and D. von der Linde are gratefully acknowledged. This work was supported by the MWF Nordrhein-Westfalen, through the Lise Meitner program.


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