Free-electron generation in laser-irradiated dielectrics

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We study the mechanisms of ultrafast free-electron generation in laser-irradiated dielectrics. The evolution of the free-electron density in the conduction band of laser-irradiated dielectrics is calculated with the recently introduced multiple rate equation. This system of rate equations 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 keeps track of the nonstationary electron energy distribution at the initial stage of ionization and provides the transition to the asymptotic avalanche regime at longer time scales. The analytic solution for the asymptotic regime yields the avalanche parameter entering the standard rate equation and the condition of its applicability. We present results on the establishment of an ionization avalanche, comparing our model with other theoretical approaches. The role of impact ionization as compared to multiphoton ionization is analyzed. A self-similarity of the fraction of impact-ionized electrons, depending only on the product of intensity and pulse duration, is revealed.

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I. INTRODUCTION

Interaction of dielectrics with ultrashort laser pulses is a broad field of fundamental theoretical and experimental investigations stimulated also by the high potential of femtosecond laser pulses in applications such as micromachining and medical surgery. When transparent solids are irradiated with laser intensities above a certain threshold, strong absorption of laser energy occurs. The increasing absorptivity is caused by the formation of a free electron gas in the conduction band of the dielectric. With the advent of ultrashort laser pulses of subpicosecond duration, a new regime of laser-matter interaction was opened where the pulse duration compares with characteristic times of microscopic collision processes within the material. Theoretical works study these microscopic processes and their influence on the free-electron distribution by solving kinetic equations such as Fokker-Planck equation or Boltzmann equation.[1–4] Numerous experimental studies investigate processes such as optical breakdown, filamentation, and Coulomb explosion, see, for example, Refs. 5–11. For all kinds of practical investigations the prebreakdown regime is of essential interest to monitor the evolution of the free-electron density and to control the laser induced damage. The temporal evolution of the free-electron density in a dielectric during ultrashort pulse laser irradiation plays therefore a fundamental role in these investigations. Usually for its description a simple rate equation is applied, though explicit kinetic calculations have shown its inadequacy on ultrashort time scales.[3,12,13]

For a long time there was a gap between the oversimplified approach of the standard rate equation and the full kinetic treatment considering the microscopic collision processes in detail. In a recent Letter[14] I introduced the multiple rate equation (MRE), which keeps track of the energy distribution of the free electrons, while maintaining the conceptual and analytic simplicity of the standard rate equation. It allows one to calculate the temporal evolution of the free-electron density on a broad range of time scales. The asymptotic solution of the multiple rate equation provides a possibility to calculate directly the avalanche parameter entering the standard rate equation and provides the condition of its applicability. Here we will present new insights into the role of the main ionization mechanisms, their mutual influence, the distinction between two fundamentally different ionization regimes, and the transition between these regimes.

In the next section we shortly resume the multiple rate equation in comparison with the commonly applied standard rate equation. Section III is devoted to concrete applications of the MRE, investigating the role of the two main ionization mechanisms in different regimes of pulse duration and laser intensity. Comparing with other theoretical approaches the MRE turns out to provide a convenient possibility to calculate the establishment of the ionization avalanche. A self-similarity is revealed concerning the fraction of impact-ionized electrons, which depends only on the product of laser intensity and pulse duration, thus on the laser fluence.

II. MODEL

Classically, the free-electron generation in dielectrics is described by a simple rate equation for the increase of the total free electron density inherent in the conduction band n_{total}:

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

This equation combines the probability of photoionization $\dot{n}_{\text{pi}}$, directly depending on the amplitude of the electric laser field E_L, with the rate of impact ionization, assumed to depend on the total free electron density.

Due to photoionization electrons are shifted from the valence band into the conduction band.[15] 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.[16,17] The avalanche coefficient α depends on the effective energy gain of the free electron in the electric laser field E_L and can be intuitively estimated[5,18] by

$$\alpha_{\text{est}} = W_{1\text{pt}}(E_L)\hbar\omega_L/\varepsilon_{\text{crit}}. \tag{2}$$

Here, $W_{1\text{pt}}(E_L)$ is the probability of one-photon intraband-absorption, $\hbar\omega_L$ the photon energy of the laser light, and $\varepsilon_{\text{crit}}$ is the critical energy for impact ionization, which is on the order of E_{gap}, the band gap between valance band and conduction band. A correction of this estimation, providing a possibility to *calculate* the avalanche parameter α was found in Ref. 14 and will be given below.

Equation (1) was proposed and verified for laser pulses in the nanosecond regime (see, for instance, Refs. 19 and 20, and references therein). Experiments measuring the optical breakdown threshold (OBT) for different pulse durations down to the femtosecond regime were analyzed by using Eq. (1), identifying the electronic avalanche as the *dominant* excitation mechanism, while multiphoton absorption only provides seed electrons for this avalanche.[1,8] However, in Ref. 10 single-shot time-resolved experiments were performed to study the free-electron density evolution in dielectrics, probing also the electron density below OBT. These experiments could only be successfully interpreted when *neglecting* the contribution of the electron avalanche, i.e., the second term in Eq. (1).

Thus, experimental studies applying Eq. (1) have lead to contradictory results. As stated in Ref. 14, the multiple rate equation (MRE) provides a possibility to clarify these controversies within the frame of a unified approach, valid on a broad range of time scales. Also earlier theoretical investigations have stated fundamental doubts whether the standard rate equation (1) is applicable in general in the subpicosecond time regime.[3,12,13] One basic assumption of Eq. (1) is that impact ionization depends directly on the total density of the free electrons. However, impact ionization needs a certain critical energy of the ionizing electron, thus this process depends also on the *energy* of a particular electron in the conduction band. While photoionization generates electrons with *low* kinetic energy in the conduction band, 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. On ultrashort time scales the shape of the electron distribution in the conduction band may change in time; then the energy-averaged total electron density n_{total} is not an adequate parameter to describe the ionization process. 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.

Defining the density n_k of electrons above $\varepsilon_{\text{crit}}$, where k will be identified with the number of photons necessary to reach $\varepsilon_{\text{crit}}$, a modified rate equation can be formulated as

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

where $\tilde{\alpha}$ represents the direct probability for impact ionization, provided that an electron with sufficient energy exists in the conduction band. $\tilde{\alpha}$ can be estimated from the corresponding collision term given for example in Ref. 3 and is, in contrast to the avalanche parameter $\alpha(E_L)$ independent of laser parameters. In the case of a stationary shape of the electron distribution in the conduction band, the fraction of high-energy electrons n_k/n_{total} is temporally constant and the modified rate equation (3) reduces to the standard rate equation (1) with $\alpha(E_L)=\tilde{\alpha}n_k/n_{\text{total}}$. For the nonstationary case, the fraction of high-energy electrons changes with time and the difference in the last term of Eq. (3) as compared to Eq. (1) is substantial.

Equation (3) follows directly from the multiple rate equation (MRE), introduced in Ref. 14. It provides a possibility to calculate the density of high-energy electrons $n_k(t)$ and thus the transient evolution of free electron density $n_{\text{total}}(t)$ also for the highly nonstationary regime on ultrashort time scales. The MRE is a comparably simple description which keeps track of the electrons energy distribution, as up to now only realized by complex kinetic approaches, while maintaining the conceptual simplicity of the standard rate equation. It is given by

$$\begin{aligned}
\dot{n}_0 &= \dot{n}_{\text{pi}} + 2\tilde{\alpha}n_k - W_{1\text{pt}}n_0,\\
\dot{n}_1 &= W_{1\text{pt}}n_0 - W_{1\text{pt}}n_1,\\
\dot{n}_2 &= W_{1\text{pt}}n_1 - W_{1\text{pt}}n_2,\\
&\vdots\\
\dot{n}_{k-1} &= W_{1\text{pt}}n_{k-2} - W_{1\text{pt}}n_{k-1},\\
\dot{n}_k &= W_{1\text{pt}}n_{k-1} - \tilde{\alpha}n_k,
\end{aligned} \tag{4}$$

with $k=\lfloor\varepsilon_{\text{crit}}/\hbar\omega_L+1\rfloor$. Here, n_j denotes the density of electrons at a discrete energy level ε_j. The $k+1$ energy levels ε_j are given by $\varepsilon_0\approx 0$, $\varepsilon_{j+1}=\varepsilon_j+\hbar\omega_L$, where $k=\lfloor\varepsilon_{\text{crit}}/\hbar\omega_L+1\rfloor$ is the integer part of $\varepsilon_{\text{crit}}/\hbar\omega_L+1$ and denotes the minimum number of photons necessary to be absorbed by an electron at $\varepsilon_0\approx 0$ to reach the critical energy for impact ionization $\varepsilon_{\text{crit}}$.

The formulation (4) of the MRE is based on a simplified view of the energy dependence of the main processes involved in the generation of free electrons in the conduction band (CB) of a dielectric, which is sketched in Fig. 1. Photoionization is assumed to generate electrons at the lower edge of the conduction band. With a certain probability $W_{1\text{pt}}$, which generally may depend on energy ε_j, such electron may absorb further single photons from the laser light and gradually reach the critical energy necessary for impact ionization. After that the electron's kinetic energy is reduced and a second electron is shifted from the valence band (VB) into the conduction band. Both electrons then have a small kinetic energy, which can be assumed to be comparable to ε_0, starting the cycle of ionization anew.

Here, relaxation processes as electron-electron relaxation and electron-phonon interaction have been neglected in order to extract the *main* effect of energy dependence of the ionization processes, leading to the failure of Eq. (1) on ul-

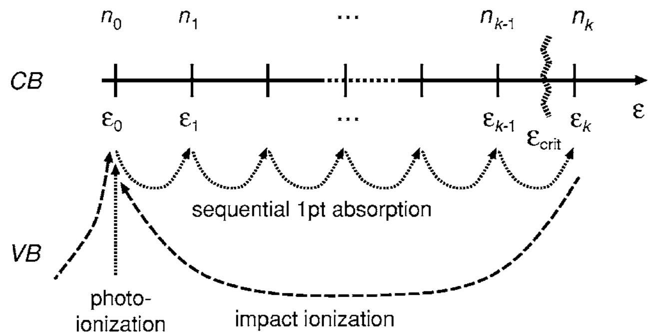

FIG. 1. Schematic view of the processes providing changes in the density and the energy, respectively, of free electrons in the conduction band of a dielectric.

trashort time scales. While electron-electron relaxation simply smears out the peaks in the electron distribution (see, for example, Fig. 3 in Ref. 3), electron-phonon interaction leads on picosecond and larger time scales to energy loss of the electron system. The reduced energy increase of the electrons during irradiation may be included qualitatively by modifying the probability $W_{1pt}(\varepsilon_j)$ to an *effective* probability of energy increase.

In general cases the probability of impact ionization $\tilde{\alpha}$ is much larger than the one-photon absorption probability W_{1pt}. For the transition $\tilde{\alpha}\rightarrow\infty$ the last equation of Eq. (4) becomes redundant and instead of the term $+2\tilde{\alpha}n_k$ in the first equation a term $+2W_{1pt}n_{k-1}$ may be used. This modification of the MRE (4) was applied in Ref. 25, it leads to a simplified numerical handling of the MRE.

As presented in Ref. 14, the MRE can be solved analytically with help of Laplace transform. For the case of $\tilde{\alpha} \gg W_{1pt}$, which is a similar but weaker assumption than the often applied "flux-doubling" model,[1,4] the solution can be found analytically. It consists of a sum of exponential functions; the largest of them takes over for long times and provides the asymptotic solution which reads

$$n_{total}(t) = \frac{\dot{n}_{pi}/W_{1pt}}{2k(\sqrt[k]{2} - 2 + \sqrt[k]{1/2})}\exp[t/t_{MRE}] \quad (5)$$

and is valid for $t \gg t_{MRE}$ with

$$t_{MRE} = [(|\sqrt[k]{2}| - 1)W_{1pt}]^{-1}. \quad (6)$$

Here, constant intensity and thus constant ionization probabilities have been assumed.

The "transition time" t_{MRE} characterizes the transition between the nonstationary regime on ultrashort time-scales and the asymptotic avalanche regime for longer times, described by Eq. (5) with the avalanche parameter

$$\alpha_{asymp} = 1/t_{MRE} = (|\sqrt[k]{2}| - 1)W_{1pt}. \quad (7)$$

The abovementioned estimated avalanche parameter α_{est}, Eq. (2), compares with the limit for $k\rightarrow\infty$ of $\alpha_{asymp} \rightarrow \ln(2)W_{1pt}/k$. Thus, α_{est} is about a factor $\ln(2)^{-1}$ larger than the *calculated* value α_{asymp}. This factor accounts in the latter value for the *doubling* of electrons in each impact ionization event.

In case of W_{1pt} depending on kinetic energy ε_j, the asymptotic behavior is similar to solution (5) with an exponent, i.e., an avalanche coefficient α, given by the largest positive real root of the polynomial

$$p(s) = (s + \tilde{\alpha})\prod_{j=1}^{k-1}[s + W_{1pt}(\varepsilon_j)] - 2\tilde{\alpha}\prod_{j=1}^{k-1}W_{1pt}(\varepsilon_j). \quad (8)$$

III. RESULTS

For the following examples, a laser with photon energy $\hbar\omega_L$=2.48 eV and electric field amplitudes up to $E_L=1\times10^{10}$ V/m was assumed, leading for a material with E_{gap}=9 eV to a critical energy of impact ionization between 13.5 and 14.5 eV,[14] hence, k=6. The probability of impact ionization $\tilde{\alpha}$ was estimated from the corresponding collision term in Ref. 3 as $\tilde{\alpha}=1\times10^{15}$ s^{-1}. The rate of photoionization $\dot{n}_{pi}$ is taken from Ref. 15, W_{1pt} is chosen to be $3.5\times10^{-7}\,E_L^2\,\mathrm{m}^2/\mathrm{V}^2\,\mathrm{s}$, which compares well with the mean value of the one-photon absorption probability for SiO_2 in Refs. 3 and 21. The effective one-photon absorption probability resulting from a fit to results of the kinetic model introduced in Ref. 3 varies less than 10% around this value for different laser intensities. The applied material parameters correspond roughly to the case of SiO_2, however, the results presented in the following are of general character. Peculiarities of quartz as ultrafast recombination in self-trapped exciton states[22,23] were neglected. Generally, recombination may be included in the multiple rate equation (4) analogously to the extension of the standard rate equation as proposed in Refs. 11 and 24. In the present work we focus on continous irradiation, thus expecting the net effect of recombination to be small. This will be the subject of a forthcoming publication.

A. Nonstationary regime

At the initial stage of ionization, the fraction of electrons with energies sufficient to perform impact ionization is expected to be small due to the non-vanishing time necessary for intraband absorption. Thus, at this stage, photoionization is the dominant ionization process. Later, the shape of the free electron distribution becomes stationary (see, for example, Fig. 3 in Ref. 3) and the probability of impact ionization, which depends on the density of high-energy electrons n_k, may be expressed through the total electron density n_{total}.

In Ref. 14, Fig. 2 we have shown the transient fraction of high-energy electrons. 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. This time scale is given by the transition time t_{MRE}, see Eq. (6), which marks the transition between the initial nonstationary regime and the asymptotic long-time regime characterized by a stationary shape of the electron distribution. The MRE model (4) thus provides a comparably simple possibility to consider not only both fundamentally different ionization regimes but also to follow the *transition* from the short-time behavior to the asymptotic avalanche regime.

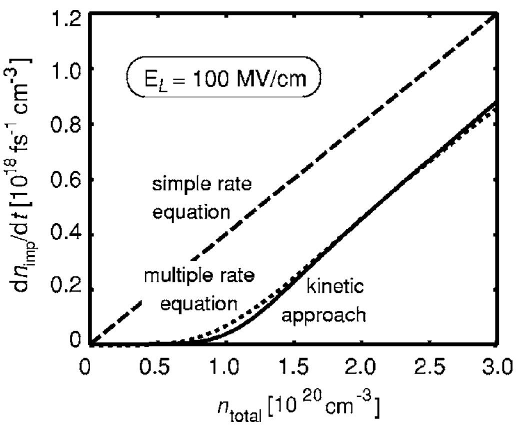

FIG. 2. Rate of impact ionization as a function of the total free electron density for constant laser field amplitude. Different models were applied: the SRE (1) (dashed line), the full kinetic model (Ref. 3) (solid line), and the MRE (4) (dotted line), respectively.

B. Establishment of the avalanche

The rate of electrons promoted in the conduction band by impact ionization $\partial n/\partial t|_{\mathrm{imp}}$ under constant laser irradiation is shown in Fig. 2 as a function of the total electron density n_{total}. While the SRE (1) predicts for constant laser intensity a linear dependence, the kinetic calculation have shown an initially depressed rate of impact ionization and only for later times a constant impact ionization rate, proportional to the total electron density (see also Ref. 3, Fig. 6). This illustrates the transition from the initial nonstationary behavior to the asymptotic avalanche regime. As Fig. 2 shows, this transition is reproduced by the MRE (4) very well. In Ref. 13 such delay of electronic avalanche was constructed with help of a steplike function. Moreover, the *initial* phase of avalanche development was reproduced through an elliptic function. In contrast, the MRE (4) yields the *complete* behavior with the transition from the initially suppressed rate of impact ionization to the asymptotic avalanche regime.

Now we are interested in the time needed to reach exponential density growth. In Ref. 1 impact ionization was studied decoupled from photoionization. The latter process was neglected and a certain distribution of "seed electrons" was assumed to feed the avalanche. Here we repeat such study applying the MRE (4) with $\dot{n}_{\mathrm{pi}}$ set to zero. Figure 3 shows the growth of total density by impact ionization assuming different initial electron distributions. We find that on a time scale of about hundred femtoseconds and less the initially assumed distribution function plays an important role. On this time scale the "time of establishment of the avalanche" strongly depends on the initially assumed conditions, thus we also may obtain an immediately established avalanche as found in Ref. 1. However, since in the real experiment the photoionization provides seed electrons at low energies at the bottom of the conduction band, a distribution of seed electrons, assuming all electrons at the lowest energy level thus contributing to n_0 seems to be the physically justified initial condition. For this case the establishment of the electron avalanche is reflected by the solid line in Fig. 3. This study confirms the transition time in the range of about 100 fs for

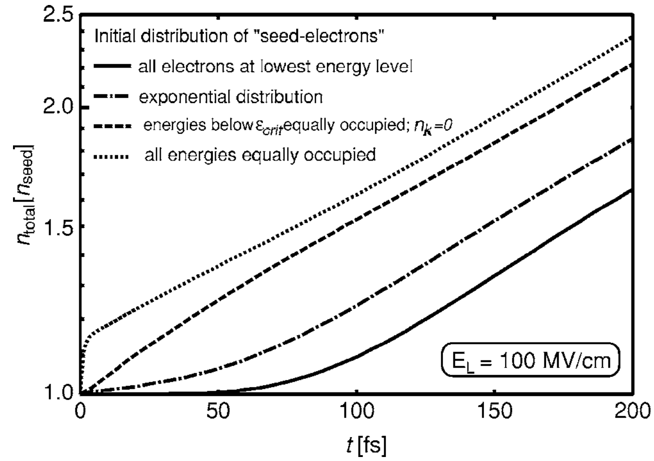

FIG. 3. Establishment of the avalanche for different initial density distributions. The time to establish the avalanche is given by the time to reach exponential density growth (linear plot) and strongly depends on the initially assumed density distribution. The physically most realistic initial distribution is the one with all electrons at the lowest energy level; the corresponding density growth is shown by the solid line. The avalanche regime is reached after about 100 fs.

the applied electric field $E_L = 100$ M V/cm, as found in Ref. 3, and the necessity of a modified approach on ultrashort time scales.

C. Total free-electron density

Figure 4 shows the transient density of free electrons under constant laser irradiation, calculated by different models. The electric laser field amplitude was assumed to be $E_L = 50$ M V/cm, resulting in a transition time $t_{\mathrm{MRE}} = 933$ fs, which is indicated as well. The density of electrons provided

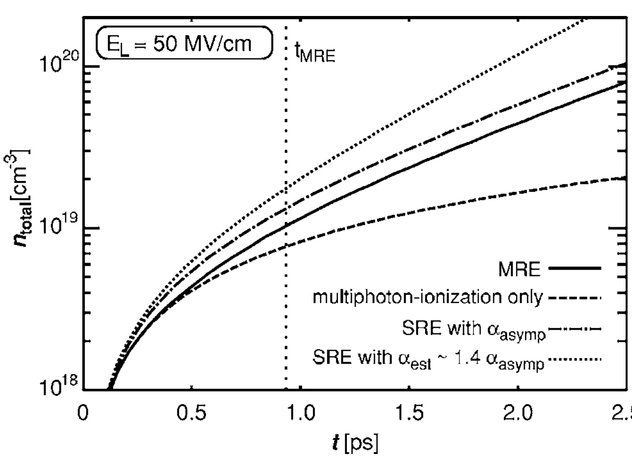

FIG. 4. Transient electron density n_{total} in the conduction band of an insulator irradiated with a laser pulse of constant electric field amplitude $E_L = 50$ MV/cm. n_{total} was calculated with different models, the multiple rate equation (MRE) (4), photoionization only and the standard rate equation (SRE) (1), with two different avalanche parameters, α_{asymp} according to Eq. (7) and α_{est} according to Eq. (2), respectively.

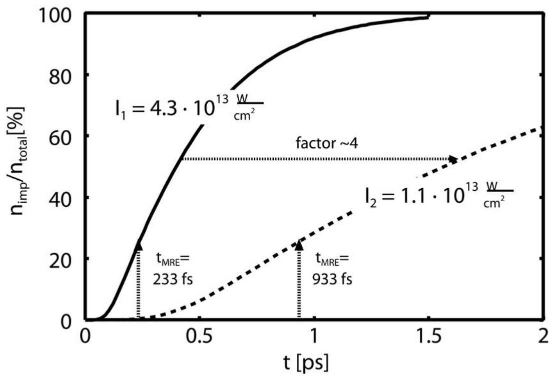

FIG. 5. Fraction of impact-ionized electrons as a function of time for constant laser irradiation of two different intensities. The curves scale with a factor of 4, as the intensities do.

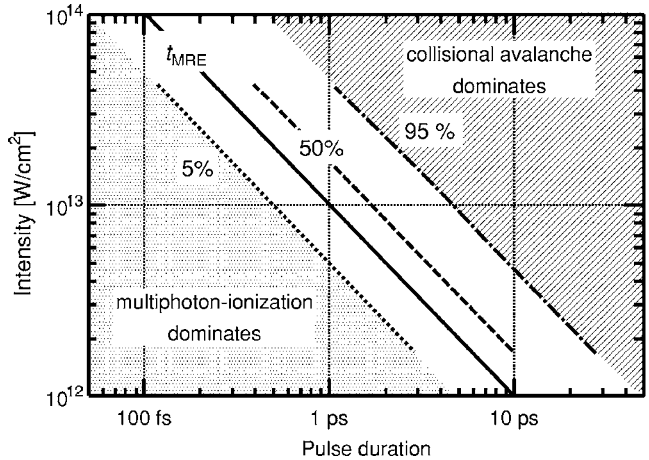

FIG. 6. Percentiles of the fraction of impact-ionized electrons $n_{\mathrm{imp}}/n_{\mathrm{total}}$ as a function of laser intensity and pulse duration. I_L-t regions where either of the ionization processes is dominating are shaded. The transition time t_{MRE} marks the transition between photoionization-dominated regime and avalanche dominated regime.

by photoionization is shown for comparison. The result of the multiple rate equation (MRE, solid line) shows that for short times $t \ll t_{\mathrm{MRE}}$ the electrons are essentially provided by photoionization. In contrast, for times $t \gg t_{\mathrm{MRE}}$ impact ionization plays an important role. If the asymptotic avalanche regime is reached, the total free electron density n_{total} grows exponentially. For this case the standard rate equation [(SRE), Eq. (1)] with $\alpha = \alpha_{\mathrm{asymp}}$ (7) provides a reasonable approximation. Figure 4 also shows the free-electron density as calculated with the SRE and the abovementioned estimated avalanche parameter α_{est} (2). Though α_{est} is only slightly larger than α_{asymp}, the free-electron density is strongly overestimated; the deviation reaches an order of magnitude after $t \approx 5t_{\mathrm{MRE}}$. Thus, the choice of the avalanche parameter α is crucial in the asymptotic long-time regime.

D. Contribution of impact ionization

A question of great interest is the fraction of impact-ionized electrons electrons provided through impact ionization $n_{\mathrm{imp}}/n_{\mathrm{total}}$ as a function of duration and intensity. Figure 5 shows the transient fraction $n_{\mathrm{imp}}/n_{\mathrm{total}}$ for a continuous laser pulse of the electric field $E_L = 100$ M V/cm, corresponding to an intensity of $I_L = I_1 = 4.3 \times 10^{13}$ W/cm^2, and for a laser pulse of $E_L = 50$ M V/cm, i.e., $I_L = I_2 = 1.1 \times 10^{13}$ W/cm^2, respectively. Here, optical parameters of SiO_2 were assumed. Concerning the higher intensity I_1, the figure shows that after a certain delay of about hundred femtoseconds the contribution of impact ionization to the total free electron density grows quickly. After about 450 fs, half of all electrons were provided by impact ionization and after 1.5 ps nearly all free electrons have been promoted to the conduction band by the process of impact ionization. It can be shown that both curves are equal: the curve for the lower intensity I_2 follows from the curve for the higher intensity $I_1 \approx 4 \cdot I_2$ by stretching the curve by a factor I_2/I_1. This scaling proceeds throughout all behavior of the fraction of impact-ionized electrons $n_{\mathrm{imp}}/n_{\mathrm{total}}$. The self-similarity can be confirmed quantitatively and is due to a self-similarity of the analytical solution of the MRE (4).

The multiple rate equation was solved in Ref. 14 for the case $\tilde{\alpha} \gg W_{1\mathrm{pt}}$ with help of the Laplace transform. Performing the transition $\tilde{\alpha} \to \infty$ and thus neglecting the last equation in equation system (4) as described above, the $\mathcal{L}$-transformed function η_{k-1} of n_{k-1} reads

$$\eta_{k-1}(s) = \frac{W_{1\mathrm{pt}}^{k-1}}{(s + W_{1\mathrm{pt}})^k - 2W_{1\mathrm{pt}}^k} \frac{\dot{n}_{\mathrm{pi}}}{s}, \qquad (9)$$

with the $k+1$ poles $s_0 = 0$, and $s_{1\cdots k} = (\sqrt[k]{2} - 1)W_{1\mathrm{pt}}$.[26,27] The full solution reads

$$n_{k-1}(t) = -\frac{\dot{n}_{\mathrm{pi}}}{W_{1\mathrm{pt}}} + \sum_{i=1\cdots k} \frac{\dot{n}_{\mathrm{pi}}}{W_{1\mathrm{pt}}} \frac{1}{K_i^*} \exp[s_i t], \qquad (10)$$

with $K_i^* = 2k(1 - \sqrt[k]{1/2})$, leading to the solution for the total free electron density

$$n_{\mathrm{total}}(t) = \frac{\dot{n}_{\mathrm{pi}}}{W_{1\mathrm{pt}}} \sum_{i=1\cdots k} \frac{1}{K_i} \exp[s_i t], \qquad (11)$$

with $K_i = 2k(\sqrt[k]{2} - 2 + \sqrt[k]{1/2})$.

Obviously, for constant intensity the density of electrons provided by photoionization is given by

$$n_{\mathrm{pi}}(t) = \dot{n}_{\mathrm{pi}} t. \qquad (12)$$

For the fraction of impact electrons follows the dependence

$$\frac{n_{\mathrm{imp}}}{n_{\mathrm{total}}} = \frac{n_{\mathrm{total}}(t) - \dot{n}_{\mathrm{pi}} t}{n_{\mathrm{total}}(t)} = \frac{\sum 1/K_i \exp[s_i t] - W_{1\mathrm{pt}} t}{\sum 1/K_i \exp[s_i t]}. \qquad (13)$$

As long as $W_{1\mathrm{pt}}$ is proportional to the intensity I_L, which is given for example for the case of Drude-like absorption, Eq. (13) depends only on the product $I_L t$. Thus, one may label the time axis in Fig. 5 anew as $I_L t$. given in 10 Ws/cm^2 for the dashed line. Thus, for constant intensity the contribution of impact ionization scales with the laser fluence. The self-similarity can be used for instance to calculate the transition time t_{MRE}, which divides the region of domination of photoionization from the avalanche dominated regime. For the given laser and material parameters the transition time may be expressed as $t_{\mathrm{MRE}} \simeq 10^{13}/I_L$ ps W/cm^2.

Figure 6 shows the fraction of impact-ionized electrons $n_{\mathrm{imp}}/n_{\mathrm{total}}$ as a function of duration and intensity. The transition time t_{MRE} marks the transition between the nonstationary short-time regime for short pulses and low intensities where photoionization is dominating and the asymptotic long-time regime for long pulses and high intensities where the impact avalanche is governing the free electron generation, respectively.

IV. SUMMARY

In summary, a model has been developed to describe the free electron density evolution in the conduction band of a dielectric under ultrashort laser irradiation. In contrast to the commonly applied simple rate equation describing only the total density of free electrons, the model presented in this article and in Ref. 14 takes into account also the *energy* of electrons in the conduction band. This is important for the ionization probability as long as the shape of electron distribution has not become stationary, i.e., in the femtosecond time regime up to picosecond time scales, depending on intensity, as the examples in this work have shown. The model leads to an ordinary differential equation system and owns much higher applicability than existing kinetic approaches. It thus provides a practical tool for such theoretical and experimental investigations where details of the collision processes are not required to know but the transient free electron density enters as a parameter.

We have studied the non-stationary regime where the fraction of free electrons with energy above critical energy for impact ionization is changing in time. The transition from the nonstationary regime to the asymptotic avalanche regime is governed by the transition time t_{MRE}, which is a function of laser intensity. The establishment of the avalanche as resulting from the MRE has been compared with other theoretical studies on this topic.[1,3,13] We have calculated the free electron density by different models and have found that for short pulse duration the photoionization essentially provides the free electrons in the conduction band while for longer pulse duration the avalanche regime takes over. In the avalanche regime the choice of the correct avalanche parameter is crucial. Finally the fraction of impact-ionized electrons has been studied in dependence on pulse duration and laser intensity which was assumed to be constant throughout irradiation. Here, a self-similarity was revealed: the contribution of impact ionization depends on the product of intensity and duration of the laser pulse, thus on the laser fluence.

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