

Interaction of femtosecond laser pulses with dielectric materials: insights from numerical modelling

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To shed light on ultra-short laser interactions, we study the laser ionization processes leading to the energy absorption and reflection. In particular, we investigate the ratio of the energy deposited to the material to the total incident energy. The absorbed energy density is studied as a function of pulse width and laser intensity. It is shown that the maximum absorption takes place at a given incident laser intensity that is considered as ablation threshold. For pulses shorter than 100 fs, only a small fraction of laser energy is deposited to the matrix, causing heating and leading to the thermal and/or mechanical modifications of the target material. We connect these results with the electronic excitation and the ionization processes leading to the changes in reflectivity and consuming electron energy. The obtained numerical results explain several recent experiments.

(Received June 14, 2009; accepted October 30, 2009)

Keywords: Femtosecond, Laser-induced breakdown, Ultrafast processes, Laser ablation

1. Introduction

The development of ultrashort laser systems has opened new possibilities for many applications in optics, photonics, electronics, and in medicine. To develop these applications, a better understanding of physical processes involved in femtosecond laser interactions is required. Therefore, many studies considered laser-induced ionization processes leading to the appearing of an optical breakdown in dielectric materials [1-5]. A detailed kinetic approach based on Boltzmann's equation was used in several papers [6,7]. Other studies employed a simplified rate equation for laser ionization [1,3-5,8]. The roles of the multi-photon ionization (MPI), the electron tunneling process, the electron-impact (avalanche) ionization were examined in these studies as a function of laser parameters. In addition, to describe the energy accumulation effects, the effects of both intrinsic and extrinsic defects (vacancies, color-centers, self-trapped excitons, dopants, impurities, etc) were outlined [4,9,10].

A further development of both industrial and medical applications of ultra-short lasers requires an improved control over the processing that is impossible without an estimation of such values as the absorbed energy, the damage and/or ablation threshold for the given laser parameters. To obtain information about these values, the electronic excitation dynamics requires more careful investigations. Therefore, we propose a model of laser interactions that accounts for the energy absorption, the ionization and the energy relaxation processes. In particular, we study the energy fraction absorbed by the target material as a function of the laser parameters

(intensity and pulse width). We connect these results with the ionization processes affecting both the reflectivity time-evolution and the energy balance.

2. Model and calculation details

To model ultra-short laser interactions with dielectric materials, we first consider the field and the electron-impact ionization processes leading to the laser energy absorption. The corresponding system of one-dimensional differential equations accounts for the multi-photon ionization process (MPI), the electron-impact (avalanche) ionization, the self-trapped exciton formation, and the plasma relaxation processes as follows [2,4,11].

$$\frac{\partial n_e}{\partial t} = \frac{(n_v - n_e)}{n_v} F + a_i n_e I + \sigma_m n_s I^m - \frac{n_e}{t_e} \quad (1)$$

$$\frac{\partial n_s}{\partial t} = -\sigma_m n_s I^m + \frac{n_e}{t_s} \quad (2)$$

$$\frac{\partial I}{\partial z} = -k\hbar\omega \frac{(n_v - n_e)}{n_v} F - \alpha_a I, \quad (3)$$

where z is the depth below the laser-irradiated surface; t is time; $n_e(t,z)$ is the number density of conduction band electrons; ω is the laser frequency; $I(t,z)$ is the laser intensity; n_v is the number density of valence band electrons in the non-excited dielectric; F is the field ionization term calculated based on the theory of Keldysh that gives an expression for both the MPI and the electron tunneling processes as a function of $I(t,z)$ [6,11]; E_g is the

energy gap; and α_a is the absorption coefficient; n_s is the number density of self-trapped excitons (STE); m is the number of photons for the STEs; $\sigma_m=10^{-93} \text{ m}^{12}\text{s}^5/\text{J}^6$ is the multi-photon cross sections for the STE; $t_s=1.5 \times 10^{-13} \text{ s}$ is the characteristic time for STEs; t_e is the electron plasma relaxation time which can range from 100 fs to 10 ps (here, $t_e=1 \text{ ps}$); and \hbar is the Plank's constant. In the present work, calculations are carried out for fused silica with parameters that are summarized in Table 1 [5,10]. The one-dimensional system is solved by using the common fourth-order Rounge-Kutta method [12]. The avalanche parameter a_i is obtained from the multiple rate equation system proposed by Rethfeld [7]. The absorption coefficient α_a is calculated based on Drude model as a function of $n_e(t,z)$ and of laser parameters. The time dependent reflectivity is given by Fresnel equations.

Table 1. Parameters used in the calculations (fused silica) [2-5,7].

Parameter	Value
Laser wavelength, λ	800 nm
Band gap, E_g	8.3 eV
Density of valence electrons, n_v	$6.6 \times 10^{28} \text{ m}^{-3}$
Free electron mobility, μ_e	$3 \times 10^{-5} \text{ m}^2/(\text{Vs})$

As soon as the conduction band electron density increases up to the critical density required for the optical breakdown, the target is transformed into a highly absorbing material. To estimate the target temperature, we therefore couple the above description of the material ionization process with the *two-temperature model* (TTM) [13]

$$C_e \frac{\partial T_e(t, z)}{\partial t} = \frac{\partial}{\partial x} K_e(T_e) \frac{\partial T_e}{\partial x} - G(T_e - T_i) + S(t, z) \quad (4)$$

$$C_l \frac{\partial T_l(t, z)}{\partial t} = \frac{\partial}{\partial x} K_l(T_l) \frac{\partial T_l}{\partial x} + G(T_e - T_l), \quad (5)$$

where $G(T_e-T_i)$ is the term that accounts for the electron-matrix energy transfer; G is the coupling parameter defined based on the electron-matrix relaxation time that is set to be 1 ps; T_e is the electron temperature; and T_i is the matrix temperature. The energy source term in the equation for the electron sub-system accounts both for the remaining electron energy after crossing the gap and for Joule heating of the electron plasma as follows [4]

$$S = \alpha_a I + (\hbar\omega - E_g) F - E_g \alpha_i n_e. \quad (6)$$

Other parameters used in the TTM, such as the electron heat capacity and the thermal conductivity, are

calculated as follows $C_e = (3/2)k_B$, and $K_e = 4k_B^2 \mu_e T_e / e$, where k_B is the Boltzmann's constant; μ_e is the free electron mobility; and e is the electron charge.

3. Results and discussion

A series of calculations are performed for a fused silica target, Gaussian laser pulse with intensity $I(t)$ at 800 nm, where I_0 is the peak laser intensity, and τ is the temporal pulse width. The effects of variations in the peak intensity and pulse width τ on ultra-short laser interactions with fused silica target are examined.

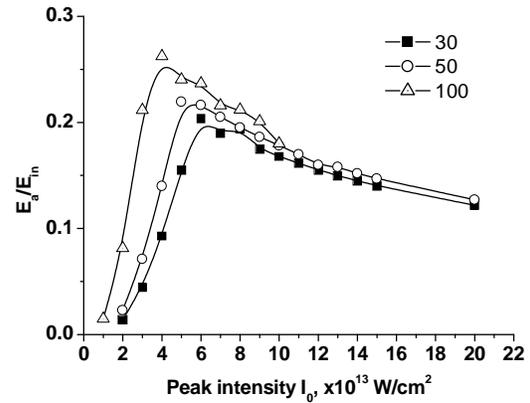


Fig. 1. Ratio of the time-integrated absorbed energy density, E_a , to that in the incident laser beam, E_{in} .

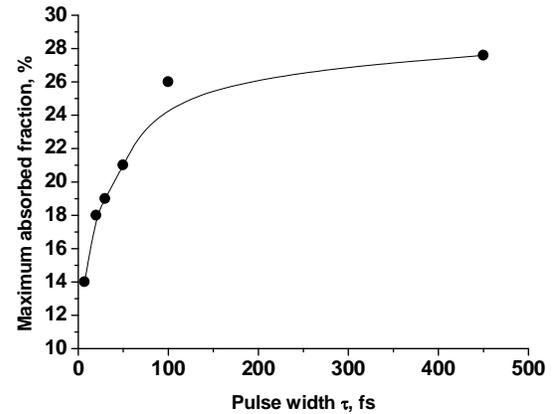


Fig. 2. Maximum possible ratio of the absorbed energy to the total incident energy, $[E_a / E_{in}]_{\max}$ as a function of pulse width.

First, we calculate the fraction of the absorbed energy density E_a to the total incident energy density $E_{in} = \int_0^{\infty} I(t) dt$ as a function of the maximum value of laser intensity I .

To analyze possible thermal effects, only the absorption by conduction band electrons (Joule heating) is considered, so that $E_a = \int_0^{\infty} \alpha_a I(t) dt$. Figure 1 shows the dependencies

obtained for three pulse durations (30, 50 and 100 fs). Interestingly, a maximum E_{max} is observed at a certain peak intensity I_{max} . For the considered pulse width range, the longer is the laser pulse, the smaller is I_{max} . This result indicates that the ablation process becomes less and less efficient with the decrease in the temporal pulse width below 100 fs.

To demonstrate the effect of pulse width on the interaction process, we plot the maximum achievable fraction of the total energy density that is absorbed by the electrons, E_{max}/E_{in} , as a function of pulse duration (Fig. 2). Note that these maximum values are achieved at different peak intensities for different pulse durations (Fig. 1). A strong decay in E_{max} is observed for $\tau < 100$ fs. In particular, only 14% of the incident energy can be absorbed by free electrons for laser pulse of 30 fs. This result is connected to the increase in reflectivity upon the optical breakdown threshold intensity (OBT) [14]. Because the experimentally measured damage threshold behaves similarly [15], a criterion based on the absorbed energy density can be used to define damage threshold. The peak value of the incident laser intensity I_0 is appropriate for the threshold definition because of the importance of the field ionization process that provides seed electrons for the following avalanche process. The calculated threshold values (Fig. 3) agree with the recent measurements of damage threshold as a function of laser pulse width [15]. For instance, our calculations give $I_0 \sim 8.0 \times 10^{13}$ W/cm² for 30 fs laser pulse, and $I_0 \sim 4.5 \times 10^{13}$ W/cm² for 100 fs laser pulse at 800 nm. The experimental values agree with our calculation results (10.0×10^{13} W/cm² for 28 fs and 3.5×10^{13} W/cm² for 100 fs).

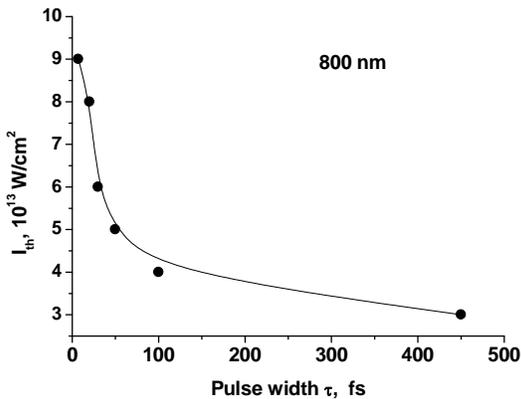


Fig. 3. Threshold laser intensity (peak value for Gaussian pulse) as a function of the temporal pulse width.

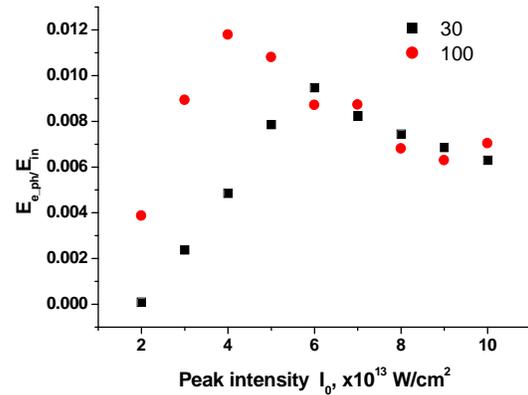


Fig. 4. Ratio of the energy deposited to the matrix to the total incident energy, E_{e-ph}/E_{in} , as a function of the laser intensity (peak value for Gaussian pulse).

It should be emphasized that only a part of the absorbed energy is transferred to the matrix/ion subsystem. The corresponding energy density in our calculations is $E_{e-ph} = \int_0^{\infty} G(T_e - T_i) dt$. The calculation

results (Fig. 4) show that the maximum energy fraction received by the matrix to the total incident energy is only around 1.2% for 100 fs and the peak intensity around the damage threshold. This value is even smaller for shorter pulses or for different laser intensities. This result is due to the energy losses for the electronic excitation and the ionization processes.

4. Summary

In summary, we have presented a study of the femtosecond laser interactions with dielectric materials. Such processes as non-linear ionization, propagation and absorption are considered in the developed numerical model. In particular, the energy absorbed by the created conduction band electrons has been examined as a function of laser intensity and pulse duration. The calculation results have demonstrated the following points:

- (i) An “optimum laser intensity” exists for a given pulse width, which maximizes the absorbed energy fraction.
- (ii) The shorter is the laser pulse, the larger is the “optimum laser intensity”.
- (iii) The fraction of the absorbed energy decays for laser pulses shorter than 100 fs.
- (iv) For laser pulses shorter than 100 fs, the maximum fraction of the energy deposited to the matrix is on the order of several percents;
- (v) Shortening of the laser pulse below 100 fs diminishes energy fraction deposited to the matrix if incident laser intensity is small enough.

These results explain the rise in the damage and ablation threshold intensities experimentally observed with the decrease in pulse width below 100 fs.

Acknowledgments

This research is supported by the French ANR-07-BLAN-0301-03 “NaNo-Morphing” program. Computer support was partly provided by the CINES of France. We are grateful to P. Lasonde and J. C. Kieffer (INRS, Varennes, Canada) for measurements of laser damage threshold and to B. Chimier (LP3 CNRS, Marseille, France) for discussions.

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