Modelling of the energy density deposition profiles of ultrashort laser pulses focused in optical media

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Abstract. The propagation of ultrashort laser pulses in dense optical media is investigated theoretically by solving numerically the nonlinear Schrödinger equation. It is shown that the maximum energy density deposition as a function of the pulse energy presents a well-defined threshold that increases with the pulse duration. As a consequence of plasma defocusing, the maximum energy density deposition is generally smaller and the size of the energy deposition zone is generally larger for shorter pulses. Nevertheless, significant values of the energy density deposition can be obtained near threshold, i.e., at lower energy than for longer pulses.

1. Introduction

Focusing intense laser pulses into optical media has attracted much interest recently due to the applications envisaged, such as the fabrication of waveguides and 3-D memories [1]. While these applications involve only small changes in the index of refraction near the focus, other applications, such as in corneal surgery, require the formation of micro-cavities through the local vaporization of the medium.

The most common approach to model the propagation of laser pulses in nonlinear media consists in solving the nonlinear Schrödinger equation (NLSE) instead of the exact Maxwell equations. Despite the approximations underlying the NLSE, it is believed that the essential physics is retained. Nevertheless, the NLSE is valid only when the electron density remains below the plasma critical density.

The NLSE has been used recently to correlate the shape of the damaged zones induced in fused silica by ultrashort laser pulses with the maximum electron density profiles [2]. However, to the best of our knowledge, no systematic investigation of the influence of the laser parameters on energy density deposition (EDD) profiles has been published yet.

In this paper we solve numerically the NLSE to study the EDD profiles in fused silica. We consider specifically this material because it is well characterized and surface damage thresholds have been measured [3]. The interest of the EDD profiles is that they can be linked directly to the local temperature reached inside the medium after electron-hole recombination (fast process), but before significant heat
diffusion (slow process). We will investigate in particular the influence of the pulse duration and pulse energy for constant focusing angle and wavelength. Although the numerical results depend on the features of the model used, we expect that the findings presented here will nonetheless provide at least a useful qualitative picture of the influence of the laser parameters on the damages induced in optical media by ultrashort laser pulses.

2. The model
In the frame of reference of the group velocity \( v_g \), the NLSE reads

\[
2i \frac{\partial E}{\partial z} + \frac{1}{k} \nabla_{\perp}^2 E - k'' \frac{\partial^2 E}{\partial t'^2} = -2k_0 n_0 |E|^2 E - i\sigma (1 + i\omega/\nu_e) n_e E - iP(|E|) E U E
\]

In this expression, \( E \) is the envelope of the electric field normalized so that \( |E|^2 \) is the laser field intensity; \( z \) is the longitudinal coordinate; \( \nabla_{\perp}^2 \) is the Laplacian operator for the transverse coordinates; \( k = k_0 n_0 \), where \( k_0 = \omega/c \) is the wave number in vacuum and \( n_0 = 1.45 \) is the linear index of refraction; \( k'' = 361 \times 10^{-30} \text{ s}^2/\text{cm} \) is the dispersion coefficient; \( n_2 = 3.54 \times 10^{-16} \text{ cm}^2/\text{W} \) is the nonlinear index of refraction; \( t' = t - z/v_g \) is the retarded time variable; \( U = 9 \text{ eV} \) is the energy gap between the valence band and the conduction band; \( P \) is the photo-ionization rate per unit volume; \( \sigma = 4\pi e^2/\nu_e m_e n_0 (1 + \omega/\nu_e) \) is the collisional absorption cross section and \( \nu_e \) is the electron-lattice collision frequency for momentum transfer, which is assumed constant here.

From Eq. (1), and the continuity equation \( \partial \Gamma / \partial t = -\partial |E|^2 / \partial z \) [3], one finds that the EDD, \( \Gamma \), is given by the equation:

\[
\frac{\partial \Gamma}{\partial t} = \sigma n_e |E|^2 + P(|E|) U
\]

For simplicity, we describe the electron density \( n_e \) by means of the single rate equation [2]

\[
\frac{\partial n_e}{\partial t} = U^{-1} \partial \Gamma / \partial t - \nu_e n_e,
\]

where \( \nu_e = 1/150 \times 10^{-15} \text{ s}^{-1} \) is the electron recombination frequency [4]. The avalanche term \( \sigma n_e |E|^2 U^{-1} \) in Eq. (3) is justified by the "flux-doubling model" [3] in the special case of a constant collision frequency.

In order to determine the collision frequency \( \nu_e \) we used Eq. (3) to fit the surface damage threshold measurements as a function of the pulse duration. The photo-ionization function used was \( P(|E|) = g|E|^{2n} \) with the coefficients \( n = 8 \) and \( g = 9.52 \times 10^{-74} \text{ cm}^{13} \cdot \text{W}^{-8} \cdot \text{s}^{-1} \) for a wavelength of 1.053 \( \mu \text{m} \) [3]. We assumed that the visible damage threshold at the target surface is determined by the EDD, \( \Gamma \), required to heat fused silica at a temperature of about 2000 K. The value of the laser field envelope \( a(t) \) is known at the target surface. Very good fits of the measurements of [3] have been obtained for \( \nu_e = 7 \times 10^{14} \text{ s}^{-1} \). This value is within the range of values reported in [5] for electrons mean energy below 5 eV.
3. Results
In this section we present the results of the numerical solution of Eqs. (1) and (3) for various laser parameters, assuming radial symmetry. All the results shown here have been obtained for a laser wavelength of 1.053 μm and for a focusing angle \( \theta_f \) (i.e., the angle of the incident cone) of 20°. This relatively small angle was used because larger angles lead to electron density exceeding the plasma critical density \( 1.01 \times 10^{21} \text{ cm}^{-3} \) for 1.053 μm) for most of the pulse energies considered, in which case the model is no longer valid. For all the results shown here, the electron density remained below the plasma critical density.

![Figure 1](image)

**Figure 1.** Contour levels of the EDD inside the target for a 10 fs (a) and a 100 fs (b) pulse. In both cases, the wavelength is 1.053 μm, the pulse energy is 0.2 μJ, and the focusing angle is 20°. The laser pulse comes from the left. The linear focus is at \( z = 75 \text{ μm} \). The dashed contour is for 100 J/cm³. A factor of ten exists between each contour level. The values increase from outside to inside.

Figure 1 shows the contour levels for the EDD in the target near the focus for two pulse durations at the same energy. One observes that the laser energy starts to be deposited before the linear focus. This is a consequence of self-focusing. Also, the lateral size of the energy deposition zone is larger for the shorter pulse. This is because the higher field intensity of the shorter pulse induces a higher photo-ionization rate, and electrons are thus created over a larger volume. (Since EDD and electron density are closely related – except for the recombination term –, only the former will be shown here.)

Figure 2a shows the maximum diameter of the EDD, defined here by the 100 J/cm³ contour, as a function of the pulse energy, for various pulse durations. One observes a threshold that increases with the pulse duration. Above the highest threshold seen in Fig. 2a, the maximum diameter decreases with the pulse duration for all pulse energies.

Despite the higher ionization rate induced by shorter pulses, the maximum EDD obtained is not higher for shorter pulses in general. As one can see in Fig. 1, the EDD exceeds \( 10^7 \text{ J/cm}^3 \) for the 100 fs pulse but not for the 10 fs pulse. Figure 2b shows the maximum EDD obtained as a function of the pulse energy and for various pulse durations. One observes that for each pulse duration there is a threshold energy below which the EDD is practically zero. This threshold increases with the pulse duration.
Moreover, the maximum EDD clearly attains higher values for larger pulse durations. This effect is a consequence of the defocusing of the pulse due to the free electrons generated by the laser pulse. The larger ionized zone induced by shorter pulses prevents the laser pulse from reaching a high intensity near the axis. For the 100 fs pulse, the maximum EDD depends weakly on the pulse energy above about 0.04 μJ. The total energy deposited is nonetheless always higher for shorter pulses.

Figure 2b also represents the temperature $T = 273 + \max(\Gamma)/C$, where $C = 1.55 \, \text{J} \cdot \text{cm}^{-3} \cdot \text{K}^{-1}$ is the heat capacity of fused silica in normal conditions. The threshold temperature corresponding to observable damage is not known. Nevertheless, assuming with other authors [2,6] that this temperature is about 1000 K (i.e., the lattice temperature corresponding to hot electrons with a density of $3 \times 10^{20} \, \text{cm}^{-3}$, after recombination [2]) our results imply that pulses much shorter than 100 fs would not induce any damage for pulse energies where longer pulses would induce damage. Moreover, our results imply that damage could be induced by 100 fs at lower energy than for longer pulses because of the lower threshold. This consequence is of importance if the smallness of the damaged zone is an important issue. Indeed, the excess energy will diffuse through thermal conduction around the energy deposition area and induce damage in a zone larger than desired. This heat diffusion is, however, a slow process and is not significant within the pulse duration for sub-picosecond pulses. As a matter of fact, the characteristic time is about $\tau_{th} = 4R^2/\kappa$ where $R$ is the radius of the energy deposition zone and $\kappa = 0.89 \, \text{cm}^2/\text{s}$ is the thermal conductance of fused silica. For $R = 1 \, \mu\text{m}$, one finds that $\tau_{th}$ is of the order of 10^{-9} \, \text{s}. This time scale is much larger than the recombination time (150 fs [4]).

Figure 2. (a) Maximum diameter of the energy deposition zone at 100 J/cm³, and (b) maximum EDD and corresponding temperature of fused silica as a function of the pulse energy, for various pulse durations. The wavelength is 1.053 μm and the focusing angle is 20°.

4. Conclusion
In this paper we have investigated the EDD profiles of laser pulses focused in fused silica by solving numerically the nonlinear Schrödinger equation. Although this theoretical framework allowed only the investigation of relatively small focusing angles, we believe that the findings of this paper would hold qualitatively for larger angles. A theoretical framework closer to the exact Maxwell equations would be necessary to provide reliable calculations in a wider range of laser parameters. Moreover, experimental
correlations between the features of the damages induced in the target and the temperature profiles near the focus would be extremely useful to benchmark the model and thus to improve our understanding of laser-induced damages in optical media.

References
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