Laser produced nanocavities in silica and sapphire: a parametric study

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Abstract. We present a model, that describes a sub-micron cavity formation in a transparent dielectric under a tight focusing of a ultra-short laser pulse. The model solves the full set of Maxwell’s equations in the three-dimensional geometry along with non-linear propagation phenomena. This allows us to initialize hydrodynamic simulations of the sub-micron cavity formation. Cavity characteristics, which depend on 3D energy release and non-linear effects, have been investigated and compared with experimental results. For this work, we want to deeply acknowledge the numerical support provided by the CEA Centre de Calcul Recherche et Technologie, whose help guaranteed the achievement of this study.

1. Introduction
Interaction of ultrashort laser pulses with transparent dielectrics is a fast growing domain of material processing, which opens a way to deposit energy inside a material so that interaction takes place in a confined geometry. Recent experiments [1] proved the possibility to focus the laser pulse into a spot of the size of a few tenths of the laser wavelength by using a microscope objective with a high numerical aperture. The energy deposited produces a strong micro explosion within the bulk. Consequently, high precision measurement of the material state after explosion, enables the tuning of equations of state in the domain of extreme parameters. In this work, electromagnetic calculations of the laser energy deposition are carried out in 2D and 3D geometries, including Kerr, dispersion, multiphoton and electron collision ionization processes. The resulting energy absorption provides the initial conditions for the 2D axially symmetric hydrodynamic calculations of the blast wave generation and the void formation.

2. Laser induced plasma discharge in dielectrics
For laser pulses duration smaller than 1 ps (shorter than the hydrodynamic response time), one can assume that atoms are at rest and only the electron dynamics needs to be considered. Modeling of sub-wavelength laser focusing requires to solve Maxwell’s equations, coupled with equations describing the non-linear response of the matter.

\[ \nabla \times \vec{E} = - \frac{\partial \vec{B}}{\partial t}, \quad \nabla \times \vec{H} = \frac{\partial \vec{D}}{\partial t} + \vec{J} + \vec{J}_{mpi}, \]

where \( \vec{J} \) is the electric current, and the last term in the right hand side is the effective current accounting for multiphoton ionization (MPI). The response of conducting electrons to the laser
electric field is considered within the Drude model
\[ \partial_t \vec{J} = -\nu_e \vec{J} + e^2 n_e \vec{E} / m_e, \]  
(2)
where \( n_e \) is the density of free electrons, and \( \nu_e = \nu_{ci} + \nu_{en} \) is the effective electron collision frequency. \( \vec{J}_{mpi} \) is obtained through a reasoning on the power loss induced by MPI, which leads to the following expression
\[ \vec{J}_{mpi} = -\vec{E} / |\vec{E}|^2 W_{ion} n_e \nu_{mpi}, \]  
(3)
where \( W_{ion} \) is ionization energy of the material (9 eV for Silica and 8.8 eV for Sapphire). Polarization is calculated using a relation including Kerr effect as below
\[ \vec{P} = \varepsilon_0 \left( \chi^{(1)} \vec{E} + \chi^{(3)} |\vec{E}|^2 \vec{E} \right) \quad \text{and} \quad \vec{D} = \varepsilon_0 \vec{E} + \frac{n_n}{n_{n0}} \vec{P}, \]
(4)
where the \( \chi^{(1,3)} \) terms are the linear and cubic dielectric susceptibilities of the material and \( \omega_0 \) is the laser frequency. Equation (4) is completed by another term describing the dispersive effects via a Sellmeier-type equation. Details about this part of the model can be found in [4]. Maxwell’s equations are complemented with a free electron density \( n_e \) evolution equation reading
\[ \partial_t n_e = \nu_{mpi} n_n + \nu_{ci} n_e - n_e / \tau_{rec}, \]
(5)
where \( n_n = n_{n0} - n_e \) is the number of neutrals. The right hand side accounts for the MPI and collisional ionization and for the radiative recombination due to the electron trapping [2]. Maxwell’s equations, coupled to density evolution equation (5), provide a self-consistent description of laser pulse interaction with dielectrics. It was solved numerically for a given set of initial conditions for the laser pulse and material properties.

3. Laser energy release
Several 2D simulations have been carried out for temporally-averaged laser pulse intensities varying from 25 to 350 TW/cm². We consider the S-polarized wave of a sinusoidal temporal shape and a convergent (110°), Gaussian transverse profile in the near field. A typical computational domain is 24 × 5 microns in the y and z directions with a 30 nm × 10 nm grid spacing for an accurate description of electric field propagation. The laser electric field is specified at the entrance plane, \( z = 0 \). A spatial distribution of absorbed energy is presented in Fig. 1 a) for the 350 TW/cm² laser intensity. Maximum laser energy is absorbed near the focal plane in a submicrometer volume. A fraction of absorbed energy in this volume depends on laser intensity as it can be seen in Fig.1 b) on the right. The energy absorption appears for the intensities above 15 TW/cm². As the laser intensity increases, the absorbed energy increases continuously and reaches the maximum of 33% around 120 TW/cm². For laser intensities higher than 350 TW/cm², energy is no longer absorbed in the focal plane but is distributed from the entrance plane to the focal plane with surfacic ablation as asymptotic behaviour. These intensities 10 – 350 TW/cm² correspond in 3D geometry to pulse energies from 10 to 100 nJ, where the cavity formation was observed in Refs. [1]. However, obtention of this well characterized energy release in the focal plane can be affected by non linear effects. A modification of the refractive index \( n \) by Kerr effect writes \( n(I) = n_0 + n_2 I \) (\( n_0 = 1.45 \) and \( n_2 = 3.54 \times 10^{16} \) cm²/W for Silica). The refractive index increases more at the center than on the edges of the beam, inducing self-focusing. For 800 nm, \( P_{cr} = 2 \) MW which can be obtained for a waist of 0.12 µm is well above the intensities used here, so we were expecting not much impact from self focusing. Simulations were nevertheless carried out for a mean intensity of 122 TW/cm² (\( E_0 = 3.5 \times 10^{10} \) V/m). The refractive index on the axis is shown in Fig. 2 a) at time \( t=120 \) fs. Kerr effect
increases the maximum index, while the electron density at the focal point is higher which can be seen by a lower index value than without Kerr effect \( n^2 = 1 + (\omega_p/\Omega)^2 \). In any case, the maximum change in the index was < 5.10^{-2} between the cases here studied, therefore confirming the neglectibility of Kerr effect in our range of intensities. Another phenomenon studied was the dispersion. For the maximum absorption coefficient presented in Fig. 1, laser energy has been kept as a constant while pulse duration \( \tau_l \) laser was adjusted from 10 fs to 200 fs. Dispersive effects are expected for small \( \tau_l \), i.e. large intensities. The dependance of absorption coefficient on the laser pulse duration is plotted in Fig. 2 b). As expected, no effects can be seen for large \( \tau_l \) whereas there is a slight bias for a small \( \tau_l \). But absorption also decreases for large intensities with dispersive effects which is due to a larger part of absorbed energy at the sample surface compared to a non dispersive case.

To conclude on 2D absorption effects, it can be shown that the dimensions of quasi-ellipsoidal absorption zone increase with laser energy by a \( I^{1/2} \) dependance (Fig. 3 on the left). Dimensions are presented in the following table for a 40 nJ absorbed energy in silica.

| 2D | 3D |
|---|---|
| e (nm) | D (nm) | L (nm) | L' (nm) | focal volume (cm³) |
| 2810 | 3680 | 1.034 10^{-13} |
| 3310 | 1.304 10^{-13} |

A similar ellipsoidal shape of the absorbing zone was found 3D simulations (Fig. 3 on the right), showing a good agreement between 2D and 3D cases. The results presented here for silica have been reproduced in sapphire with a 100 nJ laser energy (not shown here) in order to estimate the final cavity radius.
Figure 3. Left: evolution of the absorption zone surface versus laser intensity. Right: 3-D absorption map in silica with 40 nJ incident energy (laser wave is introduced in the right handside).

Figure 4. Hydrodynamic formation of cavity. Density contours in silica for 40 nJ laser energy (left) and sapphire for 100 nJ laser energy (right). Minimum density (in blue) is about 0.01 g/cm$^3$. The solid (initial) density is in red.

4. Hydrodynamic simulations

The simulations of cavity formation were performed with the hydrodynamic 2D-axisymetric two-temperature code CHIC [3]. The materials, fused silica and sapphire, have been described with SESAME tabulated equations of state (7387 for SiO$_2$ and 7411 for Sapphire). In silica and sapphire, a 40 and 100 nJ energy distribution have been respectively used as input for hydrodynamic simulations. Density contours obtained after few hundred picoseconds are presented in Fig. 4. Size of cavity estimated are similar to that measured in the experiments (175 nm / 800 nm in silica for transverse and longitudinal directions and 100 nm / 900 nm in sapphire for transverse and longitudinal directions).

5. Conclusions

We presented a model and numerical simulations describing the propagation and absorption of a short laser pulse in SiO$_2$ and sapphire, a blast wave launch and the cavity formation. The model consists of two modules - first one describes the propagation and absorption of short sub-ps laser pulses. - second one is a 2D axisymmetry hydrodynamic model implemented with an appropriate EOS.

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