Toward a new nanoLIFT transfer process

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Introduction

Electron generation and heating in dielectrics such as silica is a well known phenomena [1, 2]. The control of the deposited energy in the material allows to predict, thanks to models, its behaviour when submitted to a femtosecond, ultra-intense laser shot.

A 2D/3D self-consistent model has been developed that describes ionization process in dielectrics submitted to femtosecond laser pulses. The energy deposition is described by a full set of Maxwell’s equations in the 3D geometry. Electrons are first created either by multiphoton ionization (MPI) or tunnel ionization (TI) according to the laser intensity. Then, electrons are heated by Joule effect and they finally perform collisional ionization. The calculated absorbed energy is transferred to an hydrodynamic code that describes the shock and compression waves formation and expansion in the material.

When applied in a confined volume of water, this method leads to a jet formation at the back-surface (opposed to the irradiated surface). The aim is to improve the process of cells transfer via LIFT (Laser-Induced Forward Transfer) technique [3]. In this process, the biomaterial to be transferred is deposited on a target submitted to a laser shot, and the ejecta are collected at the backsurface on a substrate. Currently performed with nanosecond lasers, using this technique in the femtosecond regime would provide the main advantage of controlling the amount of material transferred and to proceed smaller particules.

First, multiphoton and tunnel ionization rates in silica are compared, the latter referring to Keldysh theory [4]. Characteristics of the electrons generated are described in both cases. An hydrodynamic study of water irradiated by a femtosecond laser pulse is then carried out, and jet formation mechanisms are described.

Role of tunnel effect at high intensity

When a laser beam is tightly focused inside a transparent dielectric until the beam waist reduces below the laser wavelength, the intensities reached in the focal plane are dozens of TW/cm². This leads to a swift ionization and the formation of a plasma where electrons are
heated by Joule effect. The laser beam propagation is described thanks to Maxwell’s equations, which are completed with ionization and inverse Bremsstrahlung currents. First electrons are generated by MPI, the MPI rate expressing as \( \nu_{mpi} = \sigma_n I^n \) with \( n \) the number of photons required to perform ionization, and \( \sigma_n \) the cross-section. However, at high intensities, the MPI competes with ionization by tunnel effect. The whole ionization rate given by Keldysh theory and its estimate in the tunnel regime have been computed. Keldysh ionization rates and two MPI rates with different cross-sections are plotted in Fig. 1 for a 800 nm laser shot in silica. At intensities of interest, i.e. above 30 TW/cm\(^2\), the MPI rate given by Penano (\( \sigma_6 = 9.8 \times 10^{-70} \text{s}^{-1} \left( \frac{\text{cm}^2}{\text{W}} \right)^6 \)) [5]) clearly overestimates Keldysh ionization rate. However, even if the cross-section is taken smaller (\( \sigma_6 = 3.0 \times 10^{-74} \text{s}^{-1} \left( \frac{\text{cm}^2}{\text{W}} \right)^6 \)) [6]), the MPI ionization rate is correct only in a small range of intensities - from 100 to 150 TW/cm\(^2\) in our case - due to the dependance in \( I^n \): a small variation in the intensity induces a strong variation in MPI rate. Using tunnel ionization rate would thus lead to better quantitative results.

A 2D comparison of MPI and TI estimate from Keldysh theory has been carried out. A \( \lambda = 800 \text{ nm}, \tau = 100 \text{ fs}, I = 310 \text{ TW/cm}^2 \) laser is used and the computational domain is a water area of dimensions 10 \( \mu \text{m} \) depth and 18 \( \mu \text{m} \) thickness. Figure 2 shows the electronic density and electronic temperature at the end of the laser pulse for both ionization rates.
Figure 2: Electronic density and electronic temperature on depth after a $\lambda = 800$ nm, $\tau = 100$ fs, $I = 310$ TW/cm$^2$ laser pulse in water. In solid line, tunnel ionization from Keldysh theory and in dashed line, multiphoton ionization from Penano.

Ionization in multiphoton regime begins earlier (Fig. 2.a) than in tunnel regime because of a greater ionization rate, as seen in Fig. 1. The threshold intensity for ionization is thus reached earlier with MPI and more electrons are generated. Thus, the transmitted beam is stronger in tunnel regime than in multiphoton regime because it transfers less energy to electrons at the beginning of the interaction. As a consequence, electrons created in tunnel regime are accelerated in a stronger laser field and are more energetic (Fig. 2.b).

The way electrons are ionized has a strong influence on their behaviour, and thus on the plasma formed in the material. Therefore, it is crucial to discriminate the ionization regime to model accurately the response of the material.

**Hydrodynamic motion: jet formation**

An hydrodynamic code is then used to simulate the material’s motion after the laser absorption phase. Isodensity contours on propagation axis are shown in Fig. 3 in the case of a multiphoton ionization.

At $t = 500$ ps, a shock wave has formed and propagated among the water, deforming slightly the rear surface. A cavity created by rarefaction waves emerges in the absorption zone and expands. At $t = 2.2$ ns, the cavity expansion leads to a huge deformation of the backsurface, which lasts until the cavity has no more energy to growth and begins to collapse, at $t = 5.5$ ns. During the collapse, the fluid located above the cavity is fastly absorbed in the deformed area, and a jet is formed at $t = 10$ ns. The jet velocity is $V = 376$ m$\cdot$s$^{-1}$, length is $L = 1.54$ $\mu$m and radius $R = 198$ nm, i.e. comparable to the laser beam waist $w_0 = 200$ nm.
Figure 3: Density contours on propagation axis in water at different time after a $\lambda = 800$ nm, $\tau = 100$ fs, $I = 310$ TW/cm$^2$ single laser shot coming from the left.

A similar study has been carried out for ionization by tunnel effect. In both cases, the absorption rates are around 35%. In the tunnel case, the cavity formed is more localized and thus its volumic energy is greater than in multiphoton case. The jet formed is thus faster, $V = 588$ m.s$^{-1}$, longer, $L = 2.65$ $\mu$m and the radius is smaller, $R = 180$ nm.

This method would enable to transfer nano-objects like biomolecules on substrates via LIFT technique. The choice of the focal section, directly related to the jet radius, and thus to the amount of transferred material, is the main advantage of using this technique with femtosecond lasers. This study points out that the jets characteristics are also strongly dependant on the choice of the ionization process.

References


