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Analysis of plasma-mediated ablation in aqueous tissue

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ABSTRACT

Plasma-mediated ablation using ultrafast lasers in transparent media such as aqueous tissues is studied. It is postulated that a critical seed free electron density exists due to the multiphoton ionization in order to trigger the avalanche ionization which causes ablation and during the avalanche ionization process the contribution of laser-induced photon ionization is negligible. Based on this assumption, the ablation process can be treated as two separate processes – the multiphoton and avalanche ionizations – at different time stages; so that an analytical solution to the evolution of plasma formation is obtained for the first time. The analysis is applied to plasma-mediated ablation in corneal epithelium and validated via comparison with experimental data available in the literature. The critical seed free-electron density and the time to initiate the avalanche ionization for sub-picosecond laser pulses are analyzed. It is found that the critical seed free-electron density decreases as the pulse width increases, obeying a $t_p^{-5.65}$ rule. This model is further extended to the estimation of crater size in the ablation of tissue-mimic polydimethylsiloxane (PDMS). The results match well with the available experimental measurements.

1. Introduction

Ultrashort-pulsed (USP) lasers with femtoseconds or picoseconds pulse duration have nowadays emerged as a promising tool for micro/nano-processing of various materials [1–9]. For pulses with duration width $t_p > 1$ ns, the generally accepted picture of material damage involves the heating of conduction band electrons by the incident radiation and transfer of this energy to the lattice. Damage occurs via such conventional heat deposition resulting in melting, boiling and thermal-induced fracture of materials. Because the controlling rate is that of thermal conduction through the lattice, such a heat model predicts a $t_p^{0.5}$ dependence of the threshold fluence upon pulse width t_p, in a reasonably good agreement with experiments in a variety of dielectric materials for short pulses from 100 ps to 10 ns [10]. Du et al. [11] reported that for ultrafast lasers with $t_p < 10$ ps, the damage threshold fluence is greater than the prediction from the $t_p^{0.5}$ scaling rule. Many late experimental measurements [12–14] with such ultrashort pulses confirmed the departure from the $t_p^{0.5}$ scaling rule, leading to the discovery of a new ablation category – the so-called plasma-mediated ablation [15–17].

USP laser induced plasma-mediated ablation has been described as: the interaction of a strong electromagnetic field with electrons in a condensed medium can lead to the generation of free electrons in the conduction band through multiphoton or tunnel ionization [15]. These free charges can subsequently gain sufficient kinetic energy from the electric field by inverse Bremsstrahlung (IBA) absorption to produce a large amount of free electrons – the so-called avalanche ionization [16]. The rapid ionization of the medium leads to plasma formation and a drastic increase of the local absorption coefficient which in turn gives rise to a rapid energy transfer from the radiation field to the material and results in material ablation.

It is commonly recognized that there exists a threshold for plasma-mediated ablation. An ionization rate equation [15] predicts the temporal evolution of free electrons. The threshold is determined as the free-electron density reaches to a critical value [16] – the so-called critical free-electron density. Some initial work to illustrate the underlying mechanisms of the plasma-induced ablation was conducted in pure water. It was concluded that the multiphoton ionization was most likely the pathway for generating at least one seed free electron to initiate the avalanche ionization which is predominant in the ablation process in water [15].

Extensive studies on the determination of plasma-mediated ablation thresholds in other transparent dielectrics have also been conducted [11–13] in the past two decades. To theoretically explain the discrepancy in the ultrashort pulse region, Stuart et al. [12] derived a rate equation based on production via multiphoton ionization, Joule heating, and avalanche ionization. Their model yielded a quantitative agreement with experimental measurements of the damage thresholds for fused silica at 526 and 1053 nm for pulses in a wide range from 140 fs to 1 ns. Later Tien et al. [13] developed another rate equation consisting of

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Thornber's expression for avalanche ionization and Keldysh's photoionization theory to model single-shot laser ablation, in which the discrepancy from the $t_p^{0.5}$ scaling rule was found in their damage-threshold measurements in the ablation experiments of fused silica at 800 nm in a wide pulse duration range from 10 ns down to 20 fs.

Such discrepancy has also been confirmed by Giguere et al. [18] who conducted experiments on measuring the ablation thresholds on two corneal layers – the epithelium and the stroma – with laser pulse duration from 5 ps down to 100 fs. It was found that the ablation threshold decreased rapidly with pulse durations when pulse width was greater than 1 ps. However, when the pulse width decreased to a value smaller than 1 ps, the ablation threshold did not decrease with the further decease of pulse width. Instead, a roughly constant ablation threshold was observed for pulses with duration width between 100 fs and 1 ps.

By adopting the rate equation, some authors used only one seed free electron produced via multiphoton ionization in the focal volume to initiate avalanche ionization in their calculations [15–17]. As we know, it is very easy to generate a free electron at the focus of a highly intensified laser beam when the photon energy is close to the medium band gap. However, plasma-mediated ablation occurs only when the laser irradiance is over a large threshold value. In other words, we believe that the seed free-electron density must be over a value (critical seed free-electron density) to trigger the avalanche ionization; otherwise, avalanche ionization would occur whenever a pulse irradiation is applied to a medium. Hence, one free electron is not sufficient to trigger the avalanche ionization. To answer the questions that how many seed free electrons are required to trigger avalanche ionization and when avalanche ionization occurs, we focus on establishing a theoretical model which yields an analytical solution for the required seed electron density as a function of pulse width in the present study. Consequently, the triggering time can be obtained once such a critical seed electron density is determined.

In this treatise, plasma-mediated ablation is postulated as two separate processes – the multiphoton and avalanche ionizations – during different time stages; and the rate equation accompanying each process is analytically solved. The critical seed electron density and the time when avalanche ionization occurs to induce ablation in corneal epithelium are analyzed, based on the comparison with the available experimental measurements. This analytical model in combination with the numerical modeling is employed to predict the crater size formed during the USP laser ablation of tissue-mimic polydimethylsiloxane (PDMS) for further validation of our postulation.

2. Model description

A generic rate equation consisting of multiphoton and avalanche ionizations, and diffusion and recombination losses is commonly used to predict the temporal evolution of free electrons (plasmas) in water and aqueous tissues as follows [15–17]:

$$\frac{d\rho}{dt} = \eta_{\rm mp} + \eta_{\rm ava}\rho - \eta_{\rm diff}\rho - \eta_{\rm rec}\rho^2 \tag{1}$$

where ρ is the plasma density. The first two terms on the right-hand side in the equation represent the production of free electrons through multiphoton and avalanche ionizations, respectively. The last two terms are the electron losses through diffusion and recombination, respectively.

An approximate expression for multiphoton ionization rate $\eta_{\rm mp}$ in condensed media was derived by Keldysh [19]. For the limiting condition that the optical frequency is much greater than the tunneling frequency, it follows that:

$$\eta_{\rm mp} \approx \frac{2\omega}{9\pi}\left(\frac{m\omega}{2\hbar}\right)^{3/2}\left[\frac{e^2 I(t)}{8m\Delta E\omega^2 c_0\varepsilon_0 n}\right]^k e^{2k}\Phi\left(\sqrt{2k-\frac{2\Delta E}{\hbar\omega}}\right), \tag{2}$$

in which, $\hbar$ is Dirac constant; ε_0 is the vacuum permittivity; n is the refractive index of the medium; m is the mass of electron; e (=1.6022×10^{-19} C) is an electron charge; Φ is the Dawson function; k is the number of photons required to ionize an atom or molecule $k=\left\langle \Delta E/(\hbar\omega)+1\right\rangle$, and ΔE is the band gap for ionization. The laser circular frequency is $\omega = 2\pi c_0/\lambda$, where c_0 is the speed of light in vacuum.

The avalanche ionization rate coefficient $\eta_{\rm ava}$ derived by Kennedy for ocular and aqueous media [20] is given as:

$$\eta_{\rm ava} = \frac{1}{\omega^2\tau^2+1}\left[\frac{e^2\tau}{c_0\varepsilon_0 nm\Delta E}I(t) - \frac{m\omega^2\tau}{M_m}\right] \tag{3}$$

in which, τ ($\approx$1.7 fs [21]) is the time of collision between an electron and a heavy particle, and M_m is the mass of molecule.

The laser irradiance is assumed with a Gaussian profile in both spatial and temporal domains, expressed as:

$$I(t) = (1-R)I_0\,\exp\left\{-4\ln 2[(t-t_m)/t_p]^2\right\}\exp\left(\frac{-2r^2}{w_0^2}\right), \tag{4}$$

where I_0 is the peak radiation intensity; t_p is the pulse width at half-maximum; and t_m is the time when the irradiance is in peak. The whole pulse duration is then considered as $t_d = 2t_m$. The beam radius is defined as w_0. It is noticed from the literature that the threshold for plasma-mediated ablation is defined in terms of either peak heat flux or pulse-averaged fluence. To equate these two values, it requires that:

$$\int_0^{t_d} \exp\left[\frac{-4\ \ln 2(t-t_m)^2}{t_p^2}\right]dt = t_p \tag{5}$$

The above equality gives $t_m = 0.79695t_p$. The reflectivity R is calculated by Fresnel equation at the air/material interface.

The decrease of the electron density in the focal volume by diffusion is estimated by approximating the focal volume as a cylinder with beam radius w_0 and Rayleigh length z_R, $z_R = n\pi w_0^2/\lambda$. The diffusion rate per electron is expressed as [15]:

$$\eta_{\rm diff} = \frac{\tau\Delta E}{3m}\left[\left(\frac{2.4}{w_0}\right)^2 + \left(\frac{1}{z_R}\right)^2\right]. \tag{6}$$

As for the recombination rate in the present study, it is assumed to be $\eta_{\rm rec} = 2\times10^{-9}$ cm^3/s, an empirical value obtained by Docchio [22] through measurements of the decay of plasma luminescence.

Some previous studies reported that the diffusion and recombination losses could be neglected when the pulse widths were smaller than 10 ps [13]. Therefore, the diffusion and recombination losses are assumed to be negligible in order to obtain an analytical solution of the rate equation. However, these two losses are still incorporated in the numerical modeling in this study to examine our proposed analytical model and solution.

Now based on our postulate that a critical seed free electron density must be achieved through multiphoton ionization only in order to trigger avalanche ionization and during the avalanche ionization process the contribution of multiphoton ionization is negligible, the plasma-mediated ablation process is then split into two separate ionization processes.

First, multiphoton ionization is the only mechanism to generate seed free electrons, i.e.

$$\frac{d\rho}{dt} = \eta_{\rm mp}, \qquad \text{for}\quad 0 \le t \le t_0. \tag{7}$$