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Calculation of Threshold Dependence of Laser-induced Damage upon Pulse Duration*

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Abstract A theoretical model is developed to describe the process of laser-induced damage of dielectrics with ultrashort pluses, in which the multiphoton ionization, avalanche ionization, electron-ion recombination, and the electron diffusion are taken into account. By assuming a Gaussian temporal shape of laser pulse, some numerical results of the threshold dependence on pulse duration are presented. For pulse length longer than 10ps, the numerical results agree well with the square-root relation. For pulse length of subpicosecond, our model principally explains the opposite experimental results of Du et al.'s (Appl. Phys. Lett., 64, 3071(1994) and Stuart el al.'s (Phys. Rev. Lett, 74, 2248(1995) by properly choosing the criterion of damage.

Key words: Laser-induced damage, Short laser pulse, Avalanche ionization

Laser-induced damage in otpically transparent materials has been studied extensively since the laser was invented in the 1960s[1-5]. For laser pulses longer than a few tens of picosecond, the damage mechanism is well understood. The bulk damage of defect-free dielectrics involves the heating of conduction band electrons by the incident laser field and transfer of this energy to the lattice. During the conventional heat deposition, dielectrics are melted and boiled, then damage occurs. Based on numerous experimental results, the fact that the damage threshold depends on the laser pulse width is well established. An empirical scaling law of the fluence damage threshold for longer pulses ($\tau > 10ps$) is generally accepted: [6] $F_{th} \propto \tau^{1/2}$. Meanwhile, a theoretical model which predicts this square-root relation is also set-up. [7,8]

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However, laser-induced damage by ultrashort pulses still remains as a challenging research topic because subpicosecond high power laser pulses were not available before. As a result, the mechanism of damage caused by subpicosecond pulses is only primitively understood. Until the past years, new techniques in ultrashort laser pulse generation and amplification, such as the chirped-pulse amplification (CPA), have enabled terawatt class laser systems producing subpicosecond pulses. More importantly, CPA allows one to vary the pulse width continuously from femtosecond to hundres of picosecond with a single laser system. This offers a new set of convenient opportunities to study laser-induced damage in a wide range of laser pulse widths.

In other aspect, further increase in the peak power is available from a CPA system[9], but it is now limited by optical surface damage due to the intense short pulses. Therefore, laser-induced damage, especially with short pulses, attracts much interests. In addition, various prospective applications from materials processing to biomedical technologies, such as eye surgery on ocular tissue, also need to well understand the damage mechanism of these short laser pulses and the threshold dependence on pulse duration.

Recently, Du et al.[10] investigated the laser-induced damage in fused silica employing 150 fs～7 ns, 780 nm laser pulses, and found that the threshold of damage fluence, F_{th}, increases with the pulse duration τ as $F_{th} \sim 1/\tau$ when $\tau < 1$ps. Short afterwards, Stuart et al.[11] did some similar experiments, in which they measured the damage thresholds for silica and calcium fluoride at 1053 and 526nm for pulse duration ranging from 270fs to 1 ns. But their results showed that the threshold of damage fluence still decreases with the pulse duration when $\tau < 1$ ps although it deviates from the square-root relation, which is opposite to Du et al.'s. Note that the damage detection and definitions in their experiments are different: Du et al. detected the damage through measuring the plasma emission from the sample, and they defined damage when considerable plasma emission was measured. Stuart et al. detected the damage with a Nomarski scanning electron microscope, and they defined visible permanent surface modification on the sample as damage. The differences are thought to result in their different threshold dependence on pulse duration τ for $\tau < \sim$ps.

In this paper, we develop a theoretical model to describe the process of laser-induced damage of dielectrics especially with ultrashort pulses in which the multiphoton ionization, avalanche ionization, electron-ion recombination, and the electron diffusion are taken into account. By assuming a Gaussian temporal shape of laser pulse, some numerical results of the threshold dependence on pulse duration are presented. For pulse length longer than 10 ps, the numerical results agree well with the square-root relation. For pulse lengths of subpicosecond, our model explains the opposite experimental results of Du et al.'s[10] and Stuarts et al.'s[11] by properly choosing the criterion of damage.

We use the following equation to describe the temporal behavior of the free electron density N_e.

$$\frac{dN_e(t)}{dt} = \eta(E)N_e(t) + \left(\frac{dN_e(t)}{dt}\right)_{PI} - \gamma N_e^2(t) - \delta N_e(t) \qquad (1)$$

whene $\eta(E)$ is the avalanche ionization coefficient, $\gamma = 1/(N_e(t)\tau_R)$, $\delta = 1/\tau_D$. τ_R and τ_D are the electron-ion recombination and electron diffusion time, respectively. These last two terms represent the electron losses.

The second term on the right hand of eq. (1) represents the free electron production directly by the photons of the laser radiation. In Du et al.'s and Stuart et al.'s experiments, the power density of the laser radiation is in the order of GW/cm^2$\sim$ TW/cm^2 or higher, especially for ultrashort laser pulses At this magnitude, the contribution of multiphoton ionization is nonnegligible in the processes of free electron generation Here we use the following multiphoton ionization formula[5]

$$\left(\frac{dN_e(t)}{dt}\right)_{PI} = N\,\sigma^{(n)}F^n(t) \tag{2}$$

where N is the active ion density, $\sigma^{(n)}$ is the n-photon absorption cross-section, F is the photon fluence density, respectively. We use the measured four-photon absorption cross-section $\sigma^{(4)} = 2\times 10^{-114}\,\mathrm{cm^8/s^3}$[5, 11] for the 526 nm laser field

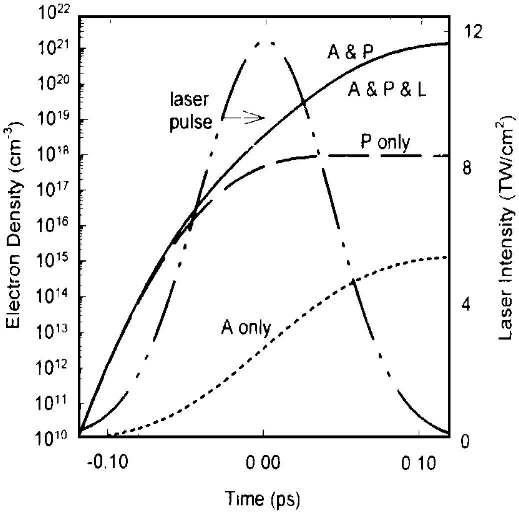
As pointed above, the power density of the laser radiation is in the order of GW/cm^2$\sim$ TW/cm^2 or higher in Du et al 's and Stuart et al 's experiments, thus the applied electric field is in the order of a few tens of MV/cm and higher, expecially for ultrashort laser pulses Under such a high field, electrons will make more than one collision during one period of the electric oscillation As a result, the electric field is essentially a dc field to those high energy electrons[10]. Hence, we take the similar way used by Du et al [10] to describe the avalanche ionization: first, we use the relationship[3] $E(\omega) = E^{dc}(1 + \omega^2\tau_c^2)^{1/2}$ corresponding the optical damage field to a dc breakdown field, where ω is the optical frequecny and τ_c is the electron collision time Second, the avalanche ionization coefficient is expressed in therms of ionization rate per unit length α with $\eta(E) = \alpha(E)\,v_{drift}$, where v_{drift} is the drift velocity of electrons When the electric field is as high as a few MV/cm, the drift velocity of free electrons is saturated and independent of the laser electric field, $v_{drift} \simeq 2\times 10^7\,\mathrm{cm/s}$ The third, we use the expression for $\alpha(E)$ derived by Thornber[12] which is applicable for all electric field strengths and essential when comparing with Du et al 's and Stuart et al 's experimental data

$$\alpha(E) = \frac{eE}{U_i}\exp\left[-\frac{E_i}{E(1+E/E_p)+E_{kT}}\right] \tag{3}$$

where U_i is the ionization threshold of the valence band electron, E_p, E_{kT} and E_i are the threshold fields for electron to overcome the deceleration effects of phonon, thermal and ionization scattering, respectively.

Setting a Gussian temporal shape of laser pulse with peak power density P and duration τ, one can determine the energy fluence F_E, photon fluence density F, and the laser electric field E and thus the corresponding dc electric field E^{dc}. Then, from eqs (1), (2), and (3) one can calculate the free electron density N_e. At the end the pulse duration, if $N_e = N_{th}$, the free electron density corresponding to the sample damage, this F_E is the damage threshold In our calculation, $E_i = 30\,\mathrm{MV/cm}$, $E_P = 3.2\,\mathrm{MV/cm}$, and $E_{kT} = 0.01\,\mathrm{MV/cm}$ are used These parameters are taken from Ref [10] (and references therein) with $U = eEl$, where U is the appropriate thermal, phonon, and ionization energy, and l is the corresponding energy relaxation length ($l_{kT} = l_p \sim 5$Å, the atomic spacing, and $l_i \simeq 30$Å).

Figure 1 and 2 are two typical runs of evolution of free electron density N_e for a 100fs and a 1 ns, 526 nm laser pulse (two-dotted dashed curve) of peak intensity 11.7 TW/cm^2 and 27.7 GW/cm^2 in fused silica, respectively. For comparison, we separte the contributions of avalanche ionization (dotted curve), and the multiphoton ionization (dashed curve). When the electron losses due to diffusion and recombination are included, the electron evolution (long-dashed curve) are also shown For high intensity laser pulses, field-induced multiphoton ionization produces the free electrons which may result in further ionization due to collision, therefore it is not necessary to invoke some arbitrary number of initial “seed” electrons Note that in Fig. 1 multiphoton ionization produces a substantial amount of free electrons, which makes some nonnegligible contributions to the sample damage. In contrast, avalanche ionization only produces a small amount of free electrons during this 100 fs pulse duration if it is taken to be the only one source of free electron generation by setting the initial electron density $N_0 = 10^{10}\mathrm{cm}^{-3}$. But if multiphoton ionization produces enough free electrons, avalanche ionization is improtant, especially at the second half of the laser pulse when the multiphoton ionization is saturated. The electron losses due to diffusion and recombination are completely negligible, hence it makes no difference for the free electron evolutions whether these losses are included or not. For 1 ns pulse duration (Fig. 2), avalanche ionization dominates the free electron generation, but contribution of multiphoton ionization is negligible except from providing the initial electrons for avalanche ionization. Because multiphoton ionization is strongly intensity dependent, the electron production takes place principally at the peak of the pulse. It is necessary to point out that the electron losses are nonnegligible for this 1 ns pulse duration; when they are included, the total electron density decreases about one order of magnitude.



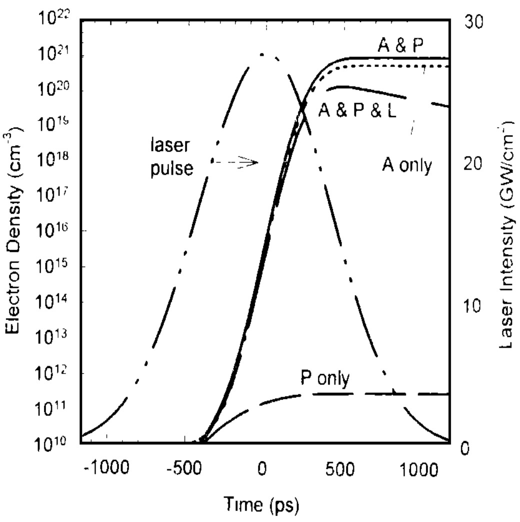


Fig. 1 Calculated evolution of free electron density N_e for a 100fs, 526 nm laser pulse (two-dotted dashed curve) of peak intensity 11.7 TW/cm^2 in fused silica. Dotted curve: only avalanche ionization is taken into account by setting the initial electron density $N_0 = 10^{10}\mathrm{cm}^{-3}$; dashed curve: only multiphoton ionization is taken into account; solid curve: both avalanche and multiphoton ionization are taken into account. When the loss term due to electron diffusion and recombination are included, the evolution of free electron density is identical to the solid curve

Fig. 2 Calculated evolution of free electron density N_e for a 1ns, 526 nm laser pulse (two-dotted dashed curve) of peak intensity 27.7 GW/cm^2 in fused silica. Dotted curve: only avalanche ionization is taken into account by setting the initial electron density $N_0 = 10^{10}\mathrm{cm}^{-3}$; short-dashed curve: only multiphoton ionization is taken into account; solid curve: both avalanche and multiphoton ionization are taken into account; long-dashed curve: all the processes of electron production and losses due to electron diffusion and recombination are included

Fig. 3 shows the calculated thresholds of damage fluences F_{th} versus laser pulse width τ for different damage conditions N_{th}. For pulses longer than 10 ps, the damage thresholds all increase with τ according to $F_{th} \propto \tau^{1/2}$, the well-known scaling. While for shorter pulses, the damage thresholds vary with the pulse duration much differently. For samll damage condition ($N_{th} = 10^{18} \mathrm{cm}^{-3}$), the threshold deviates from the square-root relation, but it still decreases with pulse duration when $\tau < 10$ ps (thick solid line). This is almost identical to the experimental results of Stuart et al [11] For bigger damage condition ($N_{th} = 10^{22} \mathrm{cm}^{-3}$), the threshold increases with pulse duration when $\tau < 2$ ps (dashed line). This is similar to the experimental results of Du et al [10] These results invoke us to conjecture that the free electron density corresponding to Du et al 's damage is smaller than that of Stuart et al 's Recalling their experiments, Du et al defined the considerable plasma emission as the sample damage , and Stuart et al defined the visible permanent modification to surface observable with a Nomarski microscope as the sample damage. In fact, considerable plasma emission can be measured only when the sample is strongly damaged. Neverthelss, surface modification can be observed with a high resolution microscope even only when slight damage occurs In addiation, for pulse duration significantly shorter than the time scale for electron energy transfer to the lattice (here subpicosecond pulse length is this situation), conduction-band electrons gain energy from the laser field much faster than they transfer to the lattice. The actual damage occurs after the pulse has passed, when this electron energy is coupled into the lattice. Du et al measured the plasma emission in situ, thus higher electron density and hence higher damage fluence are needed. In this way, our calculation principally explains the two opposite experimental results

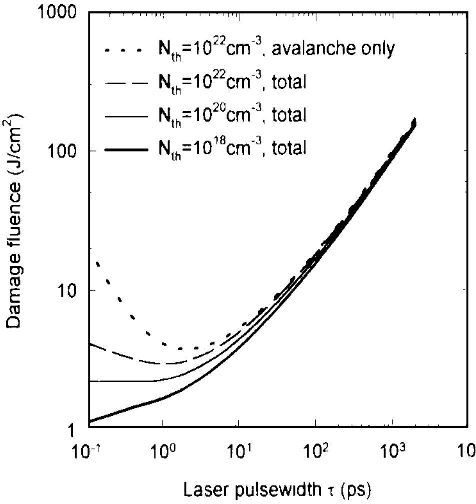


Fig. 3 Calculated thresholds of damage fluences F_{th} versus laser pulse width τ for different damage condiations N_{th}. Dotted curve: only avalanche ionization is taken into account; other three curves: all process are include. Details for shorter pulse length see text

As a reference, the threshold versus pulse duration for $N_{th} = 10^{22} \mathrm{cm}^{-3}$ when avalanche ionization is only taken into account is also shown in Fig. 3 with a dotted curve. Because contributions by multiphoton ionization are not included, the damage threshold increases with the pulsewidth more quickly. Du et al 's experimental result is close to this curve, but different from the curve of which all electron generation processes are included, as shown above. This means that multiphoton ionization almost makes no contribution to the free electron generation, which can not be explained by our present model

In conclusion, our calculations show that in the process of free electron production, the contributions of electron-ion recombination and electron diffusion are small for longer pulse width, and they are completely negligible for shorter pulse width. The avalanche ionization dominates the free electron production in the whole pulse duration for longer pulse width. But for shorter pulse width, both of multiphoton and avalanche ionization detemine the total free electron density and thus the damage threshold. More importantly our calculation results principally agree with the two opposite experiments by setting $N_{th} \sim 10^{22} \mathrm{cm}^{-3}$ (for Ref. [10]) and $\sim 10^{18} \mathrm{cm}^{-3}$ (for Ref. [11]), respectively.

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激光辐射损伤阈值与激光脉宽相互关系的模拟计算

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摘要 描述了一个超短脉冲引起的电介质激光损伤的理论模型, 这个模型考虑了多光子电离、雪崩电离、电子-离子复合和电子扩散。通过假设激光脉冲为高斯型, 数值计算得到了激光脉冲宽度与激光辐射损伤间的依赖关系。对于大于10ps 的脉冲宽度, 数值结果与平方根关系符合很好, 对于亚皮秒激光脉冲, 通过适当选择损伤的评价标准, 我们的模型解释了Du 等[10]和Stuart[11]等的相对实验结果。

关键词 激光损伤 短激光脉冲 雪崩电离

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