UCRL-JC-123302
PREPRINT

CONF-960163--21

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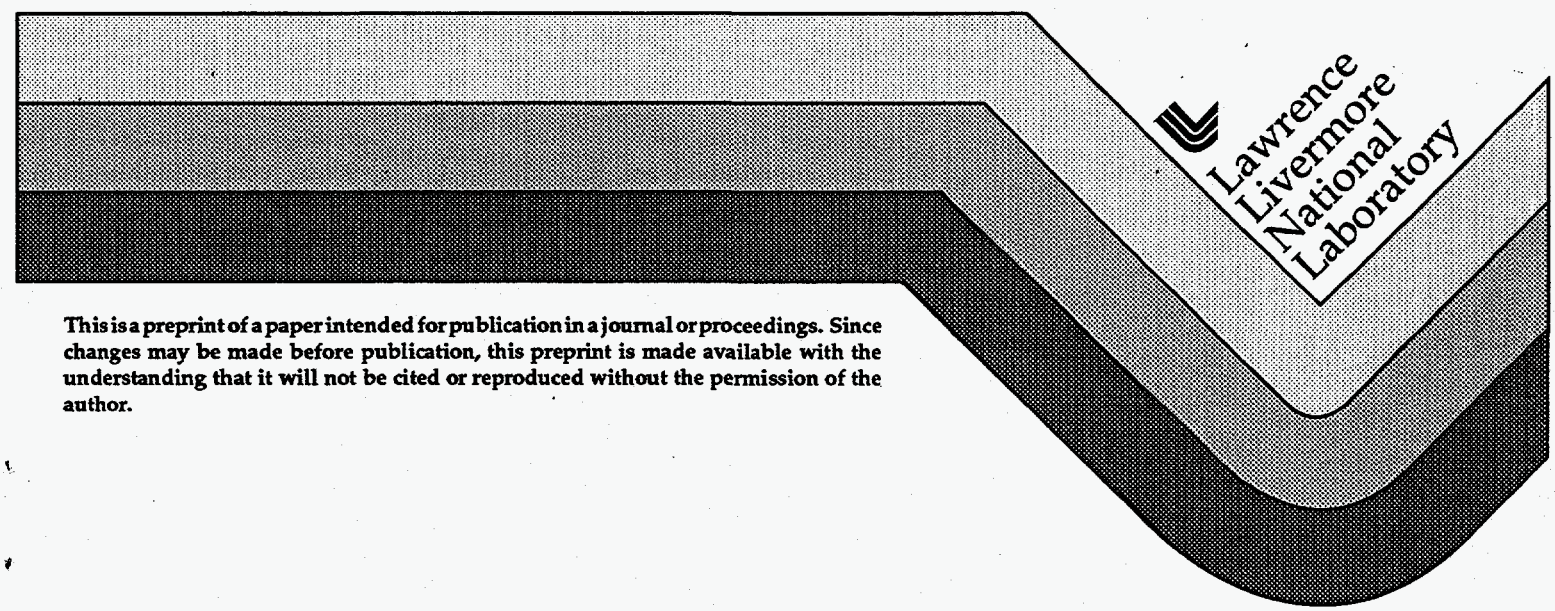
This paper was prepared for submittal to the
International Biomedical Optics '96 Symposium
San Jose, CA
January 27-February 2, 1996

February 2, 1996



Unique Aspects of Laser Energy Deposition in the fs Pulse regime*

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Biomedical Optics '96
San Jose, CA
27 January - 2 February, 1996

Ultrashort laser pulse tissue ablation has demonstrated advantages of greatly reduced required energy and collateral damage. These advantages stem directly from the fact that laser energy is absorbed nonlinearly in a time too short for significant thermal and hydrodynamic response. The high peak power and short pulse duration both have implications for practical fiber delivery systems.

keywords: ultrashort, tissue ablation, energy deposition

* Work performed under the auspices of the U.S. Department of Energy by the Lawrence Livermore National Laboratory under Contract W-7405-ENG-48

Introduction

Ultrashort (10 ps or less) laser pulse tissue ablation has demonstrated advantages of greatly reduced required energy and collateral damage[1] when compared to ns pulses which remove comparable amounts of material. Elucidation of the physical processes resulting in material removal in this regime, in particular those processes involved in the deposition of laser energy, has benefitted from studies of ultrashort laser pulse induced damage in transparent materials such as glass[2]. These studies found a distinctive change in damage morphology occurs for short pulse duration. Further, the damage energy fluence threshold depends differently on pulse duration for ultrashort pulses than it does for longer pulses, an indication of a change in fundamental mechanism. Indeed, it turns out that the physics of ultrashort pulse energy deposition is simpler than that for long pulses simply because of insufficient time for significant thermal energy transport during the deposition process. This, together with the fact that at high intensities, nonlinear absorption causes the deposition to be relatively independent of linear absorption centers, leads to a very general picture of ultrashort pulse energy deposition in transparent materials.

The advantages for tissue ablation noted above,then, can be understood to result from two novel aspects of ultrashort laser pulse energy deposition.

First, the deposition takes place so quickly that very little thermal or hydrodynamic response of the material occurs before the end of the pulse. In practical terms, this occurs for pulse durations shorter than about 10 ps. Rapid deposition simplifies the physical description since the deposition occurs independently of any thermal response. This decoupling means that less absorbed energy will be lost as waste heat

Secondly, the initial laser energy absorption is due to multiphoton ionization leading to very rapid generation of conduction electrons. This highly nonlinear process results in a very sharp ablation threshold so that material removal is highly localized and reproducible.

We have developed a rigorous theoretical description[2] of the ultrashort pulse energy absorption/avalanche process in dielectrics based on these principles. Quantitative predictions for the laser induced damage threshold in fused silica, which is well characterized physically, are in excellent agreement with observation as shown in Fig(0). This agreement gives us confidence in applying the physical picture to other insulating materials, such as biological tissue, where the details of electronic structure, etc. are not as well characterized. In the present paper, we present a simplified theoretical picture of energy deposition which allows intuitive understanding of the ultrashort pulse regime without having recourse to detailed numerical solutions of a Fokker-Planck equation.

Both high power and short pulse duration are required for ultrashort pulse tissue ablation. Thus, an optical fiber delivery system has to contend not only with laser induced damage to the fiber, but also with self-phase modulation and group velocity dispersion which tend to spread the pulse temporally. A short discussion of these effects is given.

Simple Picture of Electron Avalanche

An intense laser beam affects tissue through interactions with conduction, ie. effectively free, electrons. At high enough intensity, the field is able to directly produce promote electrons from valence states. Since the required ionization energy is typically several times the energy of a single optical photon (eg. 8 in fused silica or 6 in water for a wavelength of 1000 nm), several photons must be simultaneously absorbed for a conduction electron to be produced. Since the probability of absorbing m photons simultaneously scales as the mth power of intensity, this process only occurs at high intensity. Once such conduction electrons are present, they can be further accelerated in the electric field of the laser. Upon collision with heavy atoms and ions, the electron velocity is effectively randomized, ie. thermalized. It is possible for these heated electrons to cause further ionization if they acheive sufficient energy. A simplified rate equation model is sufficiently accurate to estimate magnitudes and aids intuitive understanding . The growth of the conduction electron density n is then described approximately by the equation

$$\frac{\partial n}{\partial t} = \beta n + S = \alpha I n + S \qquad (1)$$

where S represents the multiphoton absorption source term. Here, β is the avalanche rate which is proportional to laser intensity I provided that the intensity is larger than a threshold value. For simplicity, we assume here that I and S are independent of time. The general solution of Eq(1) is of form

$$n = \left(\frac{S}{\beta} + n_0\right) Exp[\beta t] - \frac{S}{\beta} \qquad (2)$$

where n_0 is the initial concentration. It is seen that the initial number of electrons is unimportant if it is small compared to the effective concentration S/β. If S represents multiphoton ionization (m photons), this ratio scales as the power (m-1) of intensity and the avalanche becomes independent of initial electron density for high enough intensity, ie. for short enough pulses. For example, in fused silica the effective source due to multiphoton ionization at a laser wavelength of 1000 nm is 10^{10} I^7 cm^{-3} where I is in TW/cm^2 . Thus, at I=10 TW/cm^2 , the initial electron density is immaterial provided that it is less than 10^{17} cm^{-3}. The corresponding values in typical tissue are believed to be similar. Fig.(1) displays the effective number of initial conduction electrons as a function of laser intensity

I. It is seen that for intensities of 1 TW/cm^2 or larger, any reasonable density of initial electrons due to impurities, color centers, etc. plays no role in determining the final electron density.

The avalanche coefficient $\beta = \alpha I$ can be simply understood as the rate at which energy is used to generate new electrons, ie.

$$\beta = P/U = \sigma E^2 / n U = (2\sigma / n \eta c \varepsilon_0) I \qquad (3)$$

where P is the power dissipation (Joule heating), U is the ionization energy plus the average kinetic energy of new conduction electrons, σ is the electrical conductivity, and η is the refractive index.

An essential physical quantity is the electron-lattice scattering time. We can estimate this as follows. In a noncrystalline medium, the mean free path should be on the order of an interatomic separation r. If conduction electrons have kinetic energy on the order of a few eV, the corresponding velocity is v= 10^8 cm/sec, and the scattering time is $\tau = r/v$ = a few tenths of a femtosecond, say 2×10^{-16} sec. In actuality, the electron-lattice scattering time is a function of electron energy, and this is accounted for in the full theory. Since the constant relaxation time above leads to a reasonable estimate of the electrical conductivity, it will also yield reasonable estimates of the avalanche rate and electron thermal conductivity as follows.

The dc electron mobility (drift velocity per unit electric field) is $\mu_e = e\tau/m = 0.32$ cm^2/V sec (compare 35 for Cu and 1600 for electrons in Si). The electrical conductivity is $n e \mu_e$. This gives a resistivity of $\rho = 2 \times 10^{-4}$ Ω-cm at a conduction electron density of $n = 10^{23}$ cm^{-3} .(compare 2×10^{-6} for Cu). Power is dissipated at the rate $P = \sigma E^2 = (2\sigma/\eta\, n\, c\, e_0) I \equiv \alpha U n I$ where I is the intensity in W/m^2 and η is the refractive index. U is the energy required to ionize an electron including kinetic energy; it is greater than the bandgap. The coefficient $\alpha = 2\sigma/(n\eta\, c\, \varepsilon_0 U)$ plays the fundamental role in development of the avalanche; it has the units of 1/fluence . As evident above αI represents the power dissipated per electron in units of the ionization energy. This simple picture results in estimates for α within a factor of 2 of the value resulting from detailed calculations.

The thermal and electrical conductivities of electrons are proportional (Wiedemann-Franz law) . The thermal diffusivity can thus be estimated by $v^2 \tau = kT\ \tau/m = 1$ cm^2/sec . The thermal diffusion length for a 1 ps pulse is thus on the order of 0.01 mm so no thermal diffusion takes place during a pulse of 1ps or less duration. For comparison, in Si the electron mean free path is 1000 times larger so the corresponding diffusion length is about 1 μm.

Laser Pulse Propagation

Eq.(1) describes the generation of an conduction electron plasma by the laser pulse. The presence of the plasma, of course, also affects the laser beam through reflection and absorption. Since we know the electron conductivity, we know the rate at which energy is removed from the field to produce more electrons - it is just the power density P given above. Thus, transport of electromagnetic energy is determined by

$$\frac{\partial I}{\partial z} = -\alpha U n I \equiv -a I \qquad (4)$$

where a is the local absorption coefficient (the inverse of absorption length). There is a question about what happens above the electron critical density when reflection becomes significant, but for the moment let us ignore this complication. One should solve Eqs(1)

and (4) simultaneously, ending up with n(T,z) at a time T after the pulse is over, but before thermal conduction, hydro, etc set in. One can then take U n as the deposited energy density.

Drude dielectric coefficient

The initial plasma is near solid density and exhibits a very high electron-lattice collision rate . The interaction of the laser light with the medium can be described by the usual complex Drude dielectric function (Eq.(4)). The real part of Eq.(4) describes the dielectric constant while the imaginary part (proportional to the conductivity) describes laser absorption by the conduction electrons. The example given here is for fused silica as an aid to intuition. The scattering time τ was taken as 10^{-16} sec.

The Drude dielectric coefficient is written in the form

$$\varepsilon = 1 - \frac{n}{n_c} \frac{((\omega\tau)^2 + i\omega\tau)}{1 + (\omega\tau)^2} \qquad (4)$$

in terms of the usual critical electron density n_c and light frequency ω. Of course, both the real and imaginary parts of the dielectric are linear in electron density. In the case of fused silica the term $\omega\tau$ has a value of about 0.2.

The plasma generation and laser pulse absorption can now be calculated as follows. Multiplying Eq.(1) by the energy U gives an equation for the energy density of conduction electrons. When the electron density is less than solid density, this leads to an exponential growth in the number of electrons with the average energy per electron being constant [see 2 for details]. Fig.(2) shows the evolution of electron density at the surface for a near threshold 100 fs laser pulse. The dashed line shows the pulse intensity profile for reference. The dotted curve shows the generation of conduction electrons by nonlinear multiphoton ionization. This is by far the dominant ionization mechanism,but drops off rapidly after the peak of the pulse because of the large intensity dependence. After this time, collisional ionization (inverse Bremstrahlung) due to electron acceleration in the laser electric field followed by randomizing collisions with atoms, results in a short conventional electron avalanche.

Once the material is completely ionized, further laser energy absorption results in an increase in the energy per electron. The Drude dielectric coefficient describes the propagation of the laser beam into the material. In particular, there is reflection at the surface at high electron densities. This reflection can be estimated by the Fresnel reflection formula

$$R = |(\eta-1)/(\eta+1)|^2 \qquad (5)$$

The reflection is not complete because of the very short collision time τ. The effect of reflection on the laser intensity that penetrates into the material is shown in Fig.(3) where a transmitted above threshold pulse is compared to a pulse unaffected by surface reflection. Fig.(4) compares the surface transmitted pulseshapes for several 166 fs pulses with peak intensities ranging from 16 to 20 TW/cm^2. The more intense pulses are more affected by reflection since high electron concentrations are reached sooner.

We now can use Eqs(1) and(4) to model laser energy deposition in the medium. The essential point is that since the absorption coefficient is proportional to the electron density, the laser energy will be deposited in a very thin layer once a high electron density is reached. Further increasing the laser pulse energy mainly increases the energy of this layer without greatly increasing its depth. This effect is shown in Fig.(5) where the energy deposition is shown as a function of depth into the material for the same 4 pulses shown in Fig.(4).

Implications for Delivery System

Any practical ultrashort laser pulse ablation system will require a convenient delivery system to bring the laser pulse to the targe tissue. Both articulated arms and optical fibers offer possible solutions. We discuss here some issues affecting use of optical fibers.

Although the laser energy required for ablation with ultrashort pulses is small, this peak power is quite high. Indeed as noted above, typical laser intensities are on the order of 1-10 TW/cm^2. At these intensities, several nonlinear interactions between the laser beam and the fiber can degrade or even physically damage the fiber. The Kerr effect, a nonlinear increase in refractive index which follows the spatial and temporal intensity profile, modifies the spatial and temporal properties of the laser pulse. A time dependent refractive index has the effect of broadening the spectrum of the pulse, ie. the frequency bandwidth increases. This is important for dispersion as noted below. The second effect is self focusing, in which the spatially dependent nonlinear refractive index acts like a lens. If the laser intensity in the fiber is strong enough, actual damage to the fiber core and cladding can occur[3]. Thus, one would like to use a large diameter multimode fiber to reduce the peak intensity in the fiber.

The second type of deleterious effect stems from dispersive pulse spreading due to the very short pulse duration. Optical fiber waveguides are subject to not only the usual material group velocity dispersion (different frequencies propagate with different velocities in a uniform material), but also from so-called inter-modal dispersion[4]. That is, the individual propagating modes of an optical fiber will each have a different velocity. This effect can be much larger that the material dispersion which for glass is minimal near a wavelength of 1300 nm. The minimal inter-modal dispersion occurs for a core refractive index profile having a parabolic shape. Even in this case, however, ps pulses in a typical parabolic fiber will spread in significantly in time after a propagation distance on the order of a meter. In addition to spreading in time, the relative phase shifts between propagating modes will interfere will the focusability of the light emitted from the fiber. It is for these reasons that high bandwidth telecommunications applications employ single mode (small diameter) fibers. Note that the self phase modulation mentioned above only worsens this situation since increased bandwidth leads to larger dispersion.

The two requirements of high power and short pulse duration trade off against each other when one attempts to transmit such a pulse over a length of optical fiber. We are presently investigating a number of approaches to alleviating these difficulties. In addition, other possible delivery systems such as articulated arms and hollow core fibers are being investigated.

Conclusion

Ultrashort laser pulse energy deposition in tissue is initiated by nonlinear multiphoton absorption. At intensities on the order of TW/cm^2, multiphoton ionization can produce a high density conduction electron plasma in times of 10^{-12} s or less. Further ionization is then caused by collisional ionization, ie. the acceleration of electrons in the laser field followed by collisions with ions or neutral atoms. Energy deposition takes place in a very thin surface layer. After this hot surface layer is formed, additional incident laser energy is largely reflected. Thus, there is an optimal incident energy fluence on the order of a few times the ablation threshold.

Both articulated arms and optical fibers hold promise for practical delivery of intense laser pulses.

references

[1] A.A. Orevsky, L.B. daSilva, M.D. Feit, M.E. Glinsky, B.M. Mammini, K.L. Paquette, M.D. Perry, A.M. Rubenchik, W. Small, B.C. Stuart, "Plasma mediated ablation of biological tissues with ultrashort laser pulses", Laser Tissue Interaction VI, San Jose, Ca, (6-9 Feb. 199), SPIE 2391-423 ; also see other talks this session and at this

conference: L.B. daSilva, B.C. Stuart, P. Celliers, M.D. Feit, N.J. Heredia, S.M. Herman, D.L. Mathews, M.D. Perry and A.M. Rubenchik,"Soft and hard tissue ablation using ultrashort laser pulses", SPIE 2681-26; R.A. London, D.S. Bailey, D.A. Young, M.D. Feit, A.M. Rubenchik,"Computational modeling of ultrashort pulse ablation of enamel", SPIE 2681-31

[2]B.C. Stuart, M.D. Feit, A.M. Rubenchik, B.W. Shore, and M.D. Perry, "Laser induced damage in dielectrics with nanosecond to subpicosecond pulses," Phys. Rev. Lett. **74**, 2248-51(1995); B.C. Stuart, M.D. Feit, A.M. Rubenchik, B.W. Shore, and M.D. Perry, "Nanosecond to femtosecond laser-induced breakdown in dielectrics", Phys. Rev. B (to appear Jan. 1996)

[3] H.P. Weber and W. Hodel, "High power laser transmission through optical fibers for material processing", Industrial Laser Annual Handbook 1987 edition, D. Belforte and M. Levitt,ed., SPIE vol 919, pp33-9, (Pennwell Books, Tulsa, Ok, 1987)

[4] M.D. Feit, "Minimal dispersion refractive index profiles", Appl. Opt. **18**, 2927 (1979); M.D. Feit and J.A. Fleck,Jr., "Mode properties and dispersion for two optical fiber index profiles by the propagating beam method", Appl. Opt. **19**, 3140 (1980)

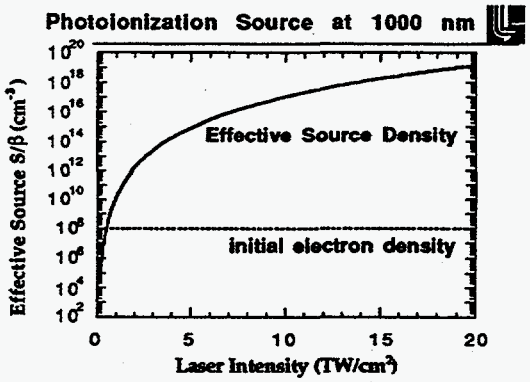

Fig. (1) - Effective density of conduction electrons due to multiphoton ionization overwhelms actual initial electron density .

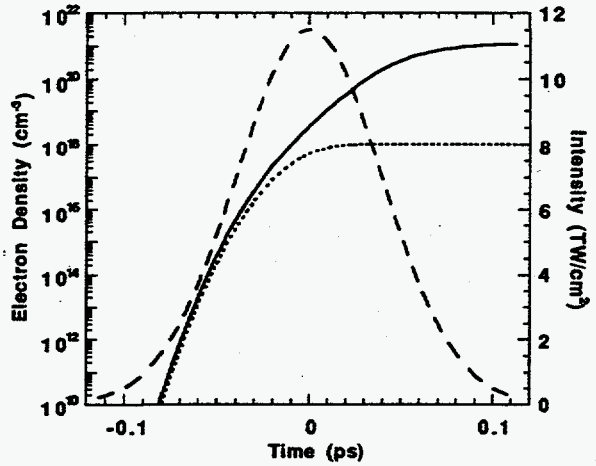

Fig. 2. Production of conduction electrons by multiphoton ionization alone (dotted curve) and by multiphoton plus collisional ionization (solid curve). The incident laser pulseshape is shown by the dashed curve for reference.

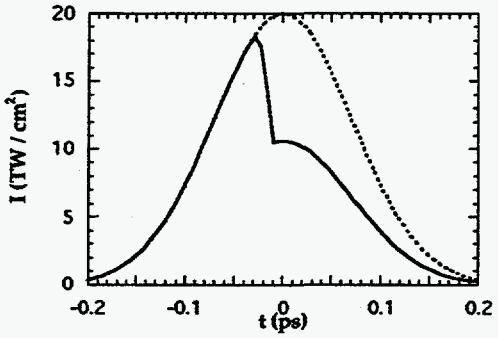

Fig. 3. Generation of high electron densities results in laser pulse reflection from the plasma. The solid curve shows the light intensity transmitted into the tissue. The dotted curve is the incident pulseshape.

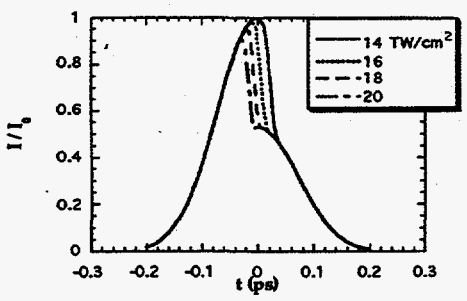

Fig. 4 . As the incident laser pulse increases in intensity from 14 to 20 TW/cm2, reflection begins sooner and a smaller fraction of the laser pulse is available to be absorbed.

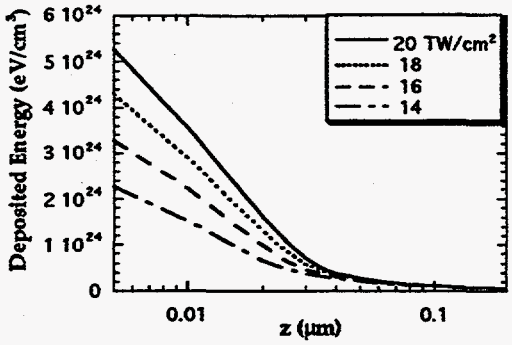

Fig. 5 Increasing the incident laser pulse intensity results in a hotter, but not much thicker energy deposition layer. Both this result and Fig. 4 imply an optimal ablation fluence.