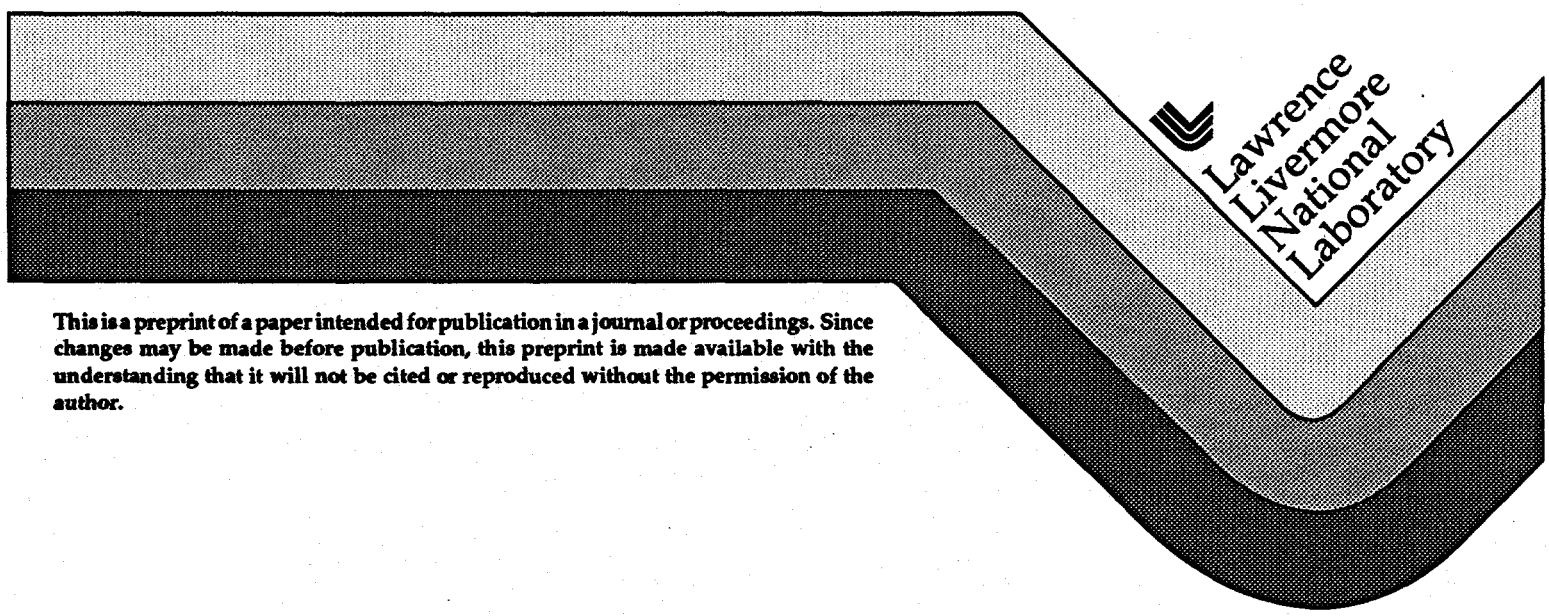
UCRL-JC-118156 Pt 2
PREPRINT

Laser-induced Damage in Dielectrics with Nanosecond to Subpicosecond Pulses II. Theory

M. D. Feit
A. M. Rubenchik
B. W. Shore
B. C. Stuart
M. D. Perry

This paper was prepared for submittal to the
Proceeding for Laser-induced Damage in
Optical Materials Conference
Boulder, CO
October 24–26, 1994

December 1994



Laser-induced damage in dielectrics with nanosecond to subpicosecond pulses: II. theory

M.D. Feit, A. M. Rubenchik, B.W. Shore, B.C. Stuart and M.D. Perry

Lawrence Livermore National Laboratory

ABSTRACT

Our extensive measurements of damage thresholds for fused silica and several fluorides (LiF, CaF, MgF and BaF) at 1053 and 526 nm for pulse durations, τ, ranging from 275 fs to 1 ns are reported elsewhere at this meeting. A theoretical model based on electron production via multiphoton ionization, Joule heating, and collisional (avalanche) ionization is in good agreement with experimental results.

Keywords: ultrashort pulses, electron avalanche, dielectric breakdown

1. INTRODUCTION

The generally accepted picture of bulk damage to defect-free dielectrics involves the heating of conduction band electrons by incident radiation and transfer of this energy to the lattice. Damage occurs via conventional heat deposition resulting in melting and boiling of the dielectric material. Because the controlling rate is that of thermal conduction through the lattice, this model predicts a $\tau^{1/2}$ dependence of the threshold fluence (energy/area) upon pulse duration τ, in reasonably good agreement with numerous experiments which have observed a τ^{α} scaling with $0.4 < \alpha < 0.5$ in a variety of dielectric materials from 100 ps to 100 ns [1].

Recently, the application of chirped-pulse amplification (CPA) [2] to solid-state lasers has enabled terawatt class systems producing subpicosecond pulses. This duration is significantly shorter than the time scale for electron energy transfer to the lattice. As a result, damage with these short pulses should exhibit a qualitative difference from that produced by longer (>100 ps) pulses.

We have developed a theoretical model in which very short intense pulses produce initial conduction band electrons by photoionization, ie. multiphoton ionization. Because the pulses are so short, Joule heating of the electrons happens too fast for significant transfer of energy from the electrons to the lattice. This heating and energy diffusion result in an electron avalanche due to impact ionization. The avalanche is described by a kinetic equation.

This model, with no adjustable parameters, is in good agreement with our observations in the short-pulse regime and over a range of laser wavelength (see accompanying paper). This model is also consistent with the observations [3] of solid density plasmas produced by ultrashort pulses.

2. THEORY OF ULTRASHORT PULSE DAMAGE THRESHOLD

2.1 Overview.

An adequate theoretical description of dielectric optical breakdown thresholds requires answering three questions. First, what are the sources of the initial electrons that initiate the avalanche? Secondly, what are the conduction electron momentum and energy scattering rates? These rates determine the rate at which laser energy can be absorbed and, thus, the avalanche rate. Thirdly, to what extent does significant heating of the lattice itself modify the scattering rates .

For ultra short pulses, a situation is encountered that is much simpler than that for long pulses. We will show the following for pulse durations less than about 10 ps. First, intensities corresponding to breakdown produce electrons via photo ionization, and these electrons initiate the avalanche. Indeed, as the sub-ps regime is reached, breakdown intensities approach the limit in which photoionization alone is capable of producing high electron densitites. Secondly, since there are no statistical fluctuations in the number of starting electrons, we will be able to define the intrinsic damage threshold of the material. The strong dependence of multiphoton rates on intensity means that the threshold becomes increasingly sharply defined for shorter durations. Thirdly, a great deal of theoretical and empirical information exists on electron scattering in silica. We are able to use an empirically based model of scattering as described below. Finally, for very short pulses, laser energy is absorbed by the electrons much faster than it can be transferred to the lattice. Since the lattice does not heat appreciably during the pulse, there is no modification of scattering rates. There is also no need to track the flow of energy into the lattice to account for thermal and mechanical stresses. The actual damage occurs after the ultrashort pulse has passed so lattice heating can be ignored for purposes of determining the damage threshold.

We take the damage threshold to be indicated by the critical electron density at which the plasma becomes reflective (10^{21} cm^{-3} for 1053 nm) since it is just below this density that the laser is strongly absorbed. Our calculations indicate the theoretical threshold is only logarithmically dependent on this choice.

2.2 Kinetic Equation

Our description of electron avalanche development is based on the solution of a kinetic equation for the electron distribution function. For insulators or other materials having a large bandgap energy U_I (i.e. $h\omega << U_I$), the heating and collision ionization of conduction electrons can be described by a Fokker-Planck equation [4,5] .

$$\frac{\partial f(\varepsilon,t)}{\partial t} + \frac{\partial}{\partial \varepsilon}\left(V(\varepsilon) f(\varepsilon,t) - D(\varepsilon) \frac{\partial f(\varepsilon,t)}{\partial \varepsilon}\right) \equiv \frac{\partial f(\varepsilon,t)}{\partial t} + \frac{\partial J(\varepsilon,t)}{\partial \varepsilon} = S(\varepsilon,t) \tag{1}$$

where

$$V(\varepsilon) = R_J(\varepsilon,t) - E_p\, \gamma(\varepsilon) = \frac{\sigma(\varepsilon) E^2(t)}{3} - E_p\, \gamma(\varepsilon) \tag{2}$$

and

$$D(\varepsilon) = \frac{2\, \sigma(\varepsilon) E^2\, \varepsilon}{3} \tag{3}$$

Here ε is the electron energy, $f(\varepsilon,\tau)\, d\varepsilon$ is the number density of electrons having kinetic energy between ε and $\varepsilon + d\varepsilon$ at time τ, R_J accounts for Joule heating of electrons in terms of the conductivity per electron σ

$$\sigma(\varepsilon) = e^2 \tau_m / m^*(1 + \omega^2 \tau_m^2), \tag{4}$$

and $\gamma(\varepsilon)$ describes the rate at which electron energy is transferred to the lattice. The quantity $1/\tau_m(\varepsilon)$ is the transport (momentum) scattering rate. Both $\tau_m(\varepsilon)$ and $\gamma(\varepsilon)$ are energy dependent, varying in fused silica by two orders of magnitude for energies in the conduction band (see Fig. 1). The current $J(\varepsilon,t)$ represents direct

heating and loss as well as an energy diffusion with coefficient $D(\varepsilon)$ which is proportional to both the conductivity and the laser intensity. The final term $S(\varepsilon,t)$ in Eq.(1) represents sources and sinks of electrons,

$$S(\varepsilon,t) = R_{imp}(\varepsilon,t) + R_{pi}(\varepsilon,t) \quad . \tag{5}$$

Impact ionization R_{imp} was included as in [5], assuming that excess kinetic energy is equally divided between the two resultant electrons. That is

$$R_{imp}(\varepsilon,t) = -\nu_i(\varepsilon) f(\varepsilon) + 4\nu_i(2\varepsilon + U_i) f(2\varepsilon + U_i) \tag{6}$$

The rate $\nu_i(\varepsilon)$was taken in the Keldysh impact ionization form [6] as $1.5\ (\varepsilon/U_I - 1)^2\ fs^{-1}$. The factor of 4 in the second term of Eq.(6) can be justified by integrating Eq.(6) over energy. This shows the net rate of electron production is simply $\int d\varepsilon\ \nu_i(\varepsilon) f(\varepsilon)$ The source term S also includes photoionization R_{pi}. At 1053 nm, we used the strong field result of Keldysh[9] corresponding to eight photon absorption. At 526 nm, we used the experimentally determined 4 photon absorption cross section $2 \times 10^{-114}\ cm^8\ sec^3$[7]. The boundary conditions for Eq.(1) are the vanishing of the distribution at $\varepsilon=\infty$ and the current at $\varepsilon=0$.

Due to the fast growth of the impact ionization rate for energy above the band gap, some researchers have replaced the source term in Eq. (6) (the full kinetic equation) by the boundary conditions.

$$f(U_i,t) = 0,\ J(0,t) = 2J(U_i,t) \tag{7}$$

These conditions imply that every electron that reaches energy U_I generates a second electron by impact ionization and leads to two electrons at energy zero. The second of these is known as the "flux doubling" condition. This formulation is advantageous if we assume exponentional growth $\exp(\beta t)$ and replace $\partial f/\partial t$ by $\beta f(\varepsilon)$. The kinetic equation is then replaced by an eigenvalue equation with β as the eigenvalue. We refer to this second formulation as the flux doubling model. The equivalence of the two formulations depends on the impact ionization rate being much larger than the rate at which the band gap energy is being absorbed. That is,

$$\sigma_{max} E^2 << U_I \nu_i(2\varepsilon + U_I)\ , \tag{8}$$

for ε small. For ultra short intense pulses, this inequality no longer holds. For example, in fused silica at 1053 nm, $\sigma_{max} E^2 = U_I \nu_i(1.5\ U_I)$ at an intensity on the order of $10\ TW/cm^2$. Thus, the equivalence of the two formulations cannot be taken for granted, but must be checked. We use this model to develop an analytic estimate of the avalanche rate which is compared to our numerical simulations of the kinetic equation.

The important physical quantities n (electron number density) and $<\varepsilon>$ (average kinetic energy per electron) are defined for the full kinetic equation by the moments

$$n = \int_0^\infty f(\varepsilon)\ d\varepsilon \tag{9}$$

$$n\ <\varepsilon> = \int_0^\infty \varepsilon\ f(\varepsilon)\ d\varepsilon \tag{10}$$

Ignoring photoionization for the time being, we see from Eqs. (1) and (6) that

$$\frac{\partial n}{\partial t} = \int_0^\infty \nu_i(\varepsilon) f(\varepsilon) d\varepsilon = <\nu> n \tag{11}$$

$$\frac{\partial (n<\varepsilon>)}{\partial t} = \int_0^\infty \left[\left(\sigma + \frac{2}{3}\varepsilon \frac{\partial \sigma}{\partial \varepsilon} \right) E^2 - E_p \gamma \right] f(\varepsilon) d\varepsilon - U_I \int_0^\infty \nu_i(\varepsilon) f(\varepsilon) d\varepsilon \tag{12}$$

$$= (<\sigma> E^2 - E_p <\gamma> - U_I <\nu>) n$$

The corresponding equations for the flux-doubling model are similar.

Aside from the derivative of σ in Eq.(12), this equation looks formally like the simple Drude theory used in [7] describing electron energy gain by Joule heating and loss by transfer to the lattice. However, the effective transport coefficients involved like

$$<\sigma> = \int_0^\infty (\sigma(\varepsilon) + \frac{2}{3}\varepsilon \frac{\partial \sigma(\varepsilon)}{\partial \varepsilon}) f(\varepsilon) d\varepsilon \Big/ \int_0^\infty f(\varepsilon) d\varepsilon \tag{13}$$

depend on averaging over the non-Maxwellian distribution function f(e) which is yet to be determined. Note that when avalanche growth occurs, <σ> of Eq.(13) will be time independent.

At high laser intensity, the energy absorbed from the laser field can not be transferred to the lattice as fast as it is being deposited in the electrons In this case, the absorbed energy is used to feed the avalanche, and the average energy per electron is high but fixed. To estimate the bounding intensity between the long and short regimes, we use Eq. (12). Initially all electrons are concentrated near the band bottom. In this case the derivative term in (12) is small and σ and γ can be evaluated at zero energy. As a result we have the condition for avalanche dominated regimes

$$\sigma(0) E^2 > E_p \gamma(0) \tag{14}$$

For parameters corresponding to fused silica, this gives I_0=80 GW/cm^2 for the limiting intensity corresponding to a pulse duration of a few hundred ps. The short pulse regime thus starts for pulse durations of less than about 10 ps.

Our calculations treat optical damage in fused silica for I >> I_0. In this case the effect of energy transfer to lattice will be small, one can consider the temperature of the lattice as a constant and, hence, consider the functions σ(ε) and $\gamma(\varepsilon)$ unchanged during the pulse.

2.3 Solutions of the Kinetic Equation

Electron scattering from various types of phonons determines the transport scattering and loss rates appearing in Eq.(1). It is currently not possible to construct a first principles theoretical model of all these interactions. The best approach combines theoretical reasoning with experimental data about interaction constants, deformation potential, etc. to construct a semi-empirical model. We used the results summarized in[8], which give a good account of electron scattering in fused silica. We digitized their data (see Fig.1) and used these rates in our calculations. We used $m^* = m_e$ and the characteristic phonon energy E_p in Eq(1) was taken as 0.033 eV. The conductivity $\sigma(\varepsilon)$ and energy diffusivity D(ε) are plotted in Fig. 2.

Numerical solution of the kinetic equation at constant laser intensity and without photoionization shows that an avalanche is established in a few fs for an intensity of 1 TW/cm^2 (see Fig. 3). The transient period before establishment of the avalanche decreases with increasing intensity since the energy diffusion increases. During the avalanche, the electron distribution grows in magnitude without changing shape, ie.

$$f(\varepsilon,t) = g(\varepsilon)\exp(\beta t) \tag{15}$$

The distribution g(ε) is stationary, but non-Maxwellian. The length of the initial transient is weakly dependent on initial conditions and decreases monotonically with intensity growth. In the flux doubling formulation, it is possible to show that for $I >> I_0$

$$\beta = pE^2 \Big/ \int_0^U \frac{d\varepsilon}{\sigma(\varepsilon)} = \alpha I \tag{16}$$

where p is a numerical factor between 0.5 and 1.0, and I is the light intensity. At 1053nm, Eq.(8) predicts α has a value between 0.0065 and 0.013 cm^2/ps GW in useful units. More conventionally, 1/α lies between. 0.08 and 0.16 J/cm^2. This sets the scale for the damage fluence as seen below. The linearity between β and intensity implied by Eq.(16) is borne out by our detailed calculations as shown in Fig. 4.

The shortness of the transient solution for constant intensity suggests that for a pulseshape I(t), we may expect a solution for the electron distribution function like $f(\varepsilon,t)=g(\varepsilon)e^{\int \beta dt}$ with $\beta = \alpha I(t)$. This supposition is tested in Fig. 5 in which we plot ln(n(t)) vs the instantaneous fluence

$$\Phi(t) = \int_{-\infty}^{t} I(t')\, dt' \tag{17}$$

for the case of a Gaussian pulse. It is evident that the linearity between β and I holds throughout nearly the entire pulse.

With the proportionality between β and I, and the exponential growth of Eq (15), the electron density can be described by

$$\frac{dn}{dt} = \beta n = \alpha I(t) n \tag{18}$$

We now reintroduce photoionization R_{pi} in the source term S of Eq.(1). This term is of form P(I)F(ε) . Here P is the photo ionization rate and F is the distribution function of the photoelectrons normalized so $\int F(\varepsilon)\, d\varepsilon = 1$. The photoionization process is sensitive to the Keldysh parameter $z = \sqrt{\frac{U_I}{U_p}}$, where U_p is the ponderomotive potential. For z>>1, which is the case here, the electron has time for many oscillations in the binding potential before being ionized. The resulting rate can be considered due to a multiphoton process .

For 526 nm light, four photon absorption is the relevant process and

$$P = \sigma_4 \left(\frac{I}{\hbar\omega} \right)^4 N_S \tag{19}$$

We used the cross section value $\sigma_4 = 2 \times 10^{-114}$ cm^8 sec^3[7]. This was measured for NaCl, but other insulators have nearly the same cross section. The quantity N_S is the solid atom density. In any case, our results are not very sensitive to the exact normalization of these rates.

For 1053 nm light, eight photon absorption crossection values were not available so we used the strong field Keldysh formula for P(I) [9]. Evaluation of the Keldysh expression leads to a result very well fit by the eight photon absorption form. We used the fit

$$P(I) = 9.5225 \times 10^{10} I^8 \text{ cm}^{-3} \text{ ps}^{-1} \tag{20}$$

where the intensity I is in TW/cm^2. This expression should be valid up to intensities on the order of 1000 TW/cm^2.

The presence of photoionization disturbs the distribution function. But if the transient time of Eqs(11,12) is small in comparison with a typical time for density increase due to photoionization, the distribution function will remain close to $g(\epsilon)$. In this case, the avalanche development is described by a simple rate equation

$$\frac{\partial n}{\partial t} = \beta(I) n + P(I) \tag{21}$$

For high photoionization rates, the rate equation can be justified as follows. The photoionization is strongly peaked at the center of the pulse. After the peak passes, photoionization becomes unimportant. The electrons produced at the peak serve as seed electrons for the avalanche. . Hence (21) can be considered as an interpolation scheme which smoothly describes the transition between the two extremes. Our numerical calculations confirm this picture. Fig. 6 compares solutions of Eqs. (1) and (21) for a 1053 nm 1 ps Gaussian pulse with peak intensity of 3.5 TW/cm^2. The close agreement between the two solutions justifies using the rate equation Eq.(21).

2.4 Determination of Damage Threshold

Fig. 7 illustrates the evolution of electron density for a 0.2 ps full 1/e width 6 TW/ cm^2 pulse. The pulse intensity and the electron density produced by photoionization alone are included for reference. Because photoionization is very intensity dependent, the electron production takes place principally at the peak of the pulse. After these "seed" electrons are produced, a small electron avalanche achieves a critical density plasma. It is important to note that the dense plasma is not produced until late in the pulse. Only this last part of the pulse experiences strong absorption or reflection. Note that we expect thresholds to be more sensitive to the pulseshape for longer pulses when the avalanche is relatively more significant.

The above numerical values all pertain to the wavelength 1053 nm. At first glance, one would expect a strong frequency dependence in the avalanche rate because of the denominator of Eq. (4). The maximum value of $\omega\tau$ at 526 nm is about 1.4. However, this value occurs at energy zero which, as remarked earlier, is relatively ineffective at determining the value of β. Instead, according to Eq(16), it is the minimal values of σ, the bottlenecks, that determine the avalanche rate. The denominator in Eq. (16) is more nearly equal to unity at these values so we do not expect a strong frequency dependence. Indeed, our numerical calculations lead to the value $\alpha = 0.0075$ cm2/ ps GW and a scale fluence of 0.26 J/cm^2.

Fig. 8 compares our theoretical values for short pulse damage thresholds in fused silica with our measured values. The agreement is very good.

3. CONCLUSION

We have described our short pulse laser induced damage measurements with a quantitative theoretical model involving no adjustable parameters. This model, taking advantage of the simplifications natural to very short pulses, gives a very good account of the measurements. This shows that we are observing intrinsic damage thresholds and, at the shorter durations, the thresholds are asymptotically limited by known multiphoton absorption rates.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under contract No. W-7405-ENG-48.

4. REFERENCES

1. J.H. Campbell, F. Rainer, M. Kozlowski, C.R. Wolfe, I. Thomas, and F. Milanovich, in *Laser Induced Damage in Optical Materials*, SPIE Vol. 1441, 444 (1990).
2. D. Strickland and G. Mourou, Opt. Comm. **56**, 219 (1985); P. Maine, D. Strickland, P. Bado, M. Pessot, and G. Mourou, IEEE J. Quantum Electron. **24**, 398 (1988).
3. B.V. Vu, O.L. Landen, and A. Szoke, Phys. Rev. E **47**, 2768 (1993).
4. A.A. Manenkov and A.M. Prokhorov, Sov. Phys. Usp. **29**, 107 (1986); L. H. Holway Jr. and D. W. Fradin, J. Appl. Phys. **46,** 279 (1974).
5. M. Sparks, D. L. Mills, R. Warren, T. Holstein, A. A. Maradudin, L. J. Sham, E. Loh Jr., and D. F. King, Phys. Rev. B **24,** 3519 (1981).
6. B.K. Ridley, *Quantum Processes in Semiconductors* (Clarendon, Oxford, 1993).
7. S. C. Jones, P. Braunlich, R. T. Casper, X.-A. Shen, and P. Kelly, Opt. Eng. **28,** 1039 (1989).
8. D. Arnold, E. Cartier and D. J. DiMaria, Phys. Rev. A **45,** 1477 (1992).
9. L.V. Keldysh, Sov. Phys., JETP **20**, 1307 (1965).