

CONF-9505264-25-REV.

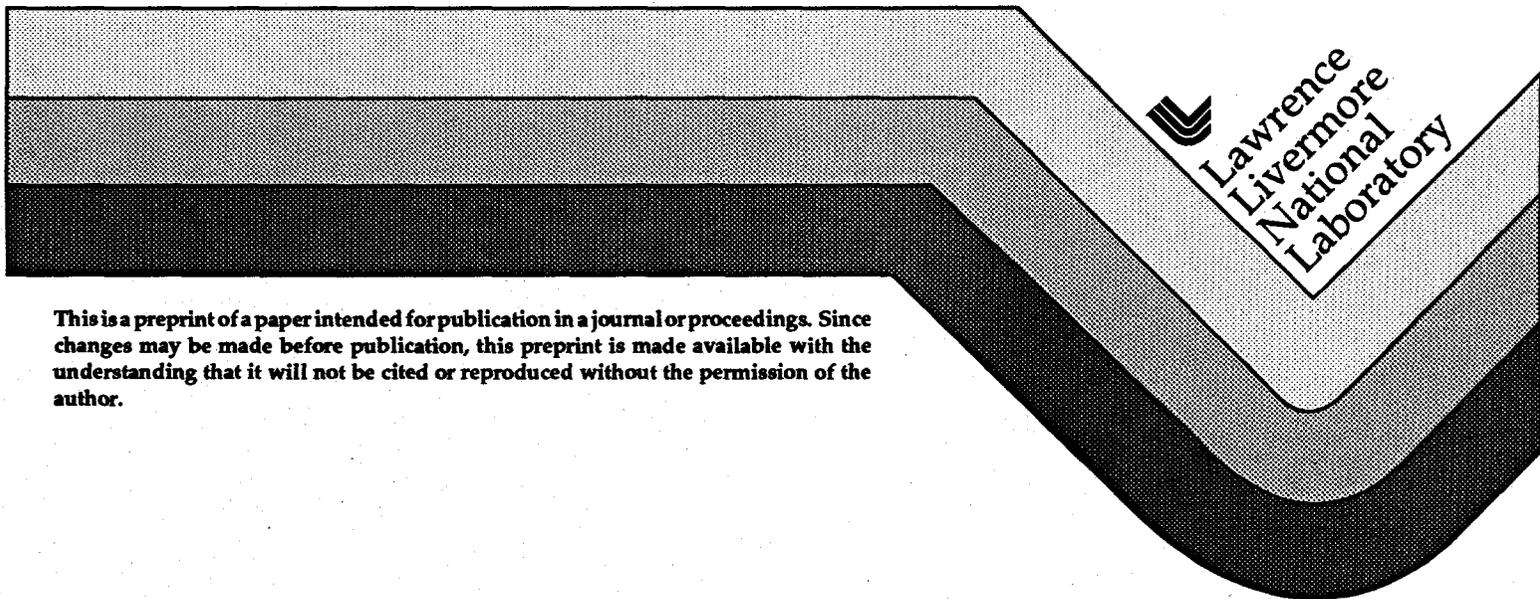
UCRL-JC-120187 Rev 1  
PREPRINT

## Laser Induced Damage in Multilayer Dielectric Gratings Due to Ultrashort Laser Pulses

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

This paper was prepared for submittal to the  
1st Annual International Conference on Solid-State Lasers  
for Application to Inertial Confinement Fusion  
Monterey, CA  
May 30-June 2, 1995

July 11, 1995



This is a preprint of a paper intended for publication in a journal or proceedings. Since changes may be made before publication, this preprint is made available with the understanding that it will not be cited or reproduced without the permission of the author.

DISTRIBUTION OF THIS DOCUMENT IS UNLIMITED

MASTER

#### DISCLAIMER

This document was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor the University of California nor any of their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or usefulness of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or the University of California. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or the University of California, and shall not be used for advertising or product endorsement purposes.

## **DISCLAIMER**

**Portions of this document may be illegible in electronic image products. Images are produced from the best available original document.**

## Laser induced damage in multilayer dielectric gratings due to ultrashort laser pulses

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

*Lawrence Livermore National Laboratory, PO Box 808, L-493, Livermore CA 94550*

### ABSTRACT

Chirped pulse amplification is increasingly used to produce intense ultrashort laser pulses. When high-efficiency gratings are the dispersive element, as in the LLNL Petawatt laser, their susceptibility to laser induced damage constitutes a limitation on the peak intensities that can be reached. To obtain robust gratings, it is necessary to understand the causes of short-pulse damage, and to recognize the range of design options for high efficiency gratings. Metal gratings owe their high efficiency to their high conductivity. To avoid the inevitable light absorption that accompanies conductivity, we have developed designs for high efficiency reflection gratings that use only transparent dielectric materials. These combine the reflectivity of a multilayer dielectric stack with a diffraction grating. We report here our present understanding of short-pulse laser induced damage, as it applies to dielectric gratings.

**Keywords:** Gratings, damage

### 1. INTRODUCTION

The intensity available from a short pulse laser system is limited by a number of factors. For chirped pulse amplification, using grating-pair pulse compression<sup>1,2</sup>, an important limitation is set by the ability of the gratings to withstand high fields. The conductivity of metals acts to exclude electric fields from the interior, but because the conductivity is not infinite, some field penetrates and is absorbed. This absorbed energy limits the allowable fluence onto a metal grating. Our high efficiency multilayer dielectric gratings<sup>3</sup> offer an attractive alternative to traditional metallic gratings. Here we report on theory and experiments pertaining to their damage thresholds.

We follow tradition and quantify damage by the fluence ( $\text{J}/\text{cm}^2$ ) needed to produce any change visible under a microscope. The morphology of the damage may take many forms, including fracturing, melting and recrystallization, and ablation. The nature of the damage can vary dramatically, depending on the pulse duration and the presence of surface or subsurface damage-prone sites.

### 2. EXPERIMENTAL METHOD

For damage testing we used laser pulses generated by a 1053-nm Ti:sapphire CPA system<sup>4-6</sup> in which seed pulses of 100 fs from a Kerr-lens mode-locked, Ti:sapphire oscillator were stretched to 1 ns in a four-pass, single-grating (1740 line/mm) pulse stretcher and then amplified by nearly  $10^9$ , to the 6-mJ range, in the TEM<sub>00</sub> stable cavity mode of a linear regenerative amplifier. Further amplification to the 60-mJ level was achieved in a Ti:sapphire ring regenerative amplifier. This system operated at 10 Hz.

After amplification, we compressed the pulses in a four-pass, single-grating compressor. By varying the dispersive path length of the compressor, we obtained pulses of continuously adjustable duration from 0.4 to 1 ns (all reported pulsewidths are intensity full-width at half-maximum). For these damage measurements, we compressed a near-Gaussian spectral profile to obtain temporally smooth output pulses. This allowed us easily to relate the time evolution of the pulse intensity to the measured fluence.

### 3. LASER INDUCED DAMAGE

Damage produced by pulsed lasers in transparent dielectrics begins with the conversion of electromagnetic energy into kinetic energy of electrons. Although the damage is manifest in lattice changes, the coupling with the laser pulse occurs through electrons. To produce damage, energy deposited with electrons must be transformed into structural changes of the lattice. Our recent experiments, supported by theoretical developments, have shown the importance of pulse duration as a determining factor in laser induced damage<sup>5,6</sup>. Our work shows that damage induced in dielectrics by ultrashort pulses (shorter than a few tens of picoseconds) is fundamentally different from that occurring for longer pulses (see also reference<sup>7</sup>).

### 4. LONG PULSE DAMAGE

Damage produced in transparent dielectrics by laser pulses with duration  $\tau$  longer than a few tens of picoseconds may take many forms, including

- Fracturing, caused by stresses due to heating
- Melting, possibly followed by recrystallization
- Ablation or evaporation.

Long pulse damage has the following general characteristics:

- The damage is very strongly dependent on the presence of defects and irregularities
- The damage has a statistical nature: measured thresholds show fluctuations
- The damage fluence is proportional roughly to the square root of the pulse duration.

### 5. SHORT PULSE DAMAGE

Whereas long pulse damage can take many forms, short pulse damage is more sharply defined and has less varied morphology:

- Short pulse damage has the appearance of ablation
- The ablation is not accompanied by collateral damage to surrounding regions

Damage by short pulses has the following general characteristics

- The damage has a very sharply defined threshold. We found that 10,000 subpicosecond pulses produced no damage when the fluence was reduced by only 2% below our experimentally determined threshold
- The damage fluence deviates markedly from the square-root dependence on pulse duration.

The damage threshold becomes increasingly well defined for short pulses, ie. it becomes independent of probabilistic factors such as distribution and concentration of defects. We have found that the short-pulse damage threshold is independent of laser spot size; others\ have found that femtosecond ablation rates in dielectrics are independent of laser spot size.

## 6. BASIC CONCEPTS OF SHORT PULSE DAMAGE

When the pulse duration becomes sufficiently short (less than 20 ps), there is insufficient time for electron energy to be converted into lattice energy. Energy absorbed by electrons remains as electron energy. The onset of damage from short pulses in pure transparent materials is simpler than that from long pulses, for the following reasons.

1. At the intensities for which short pulse damage occurs, multiphoton ionization is capable of supplying conduction electrons. These photoelectrons outnumber any electrons initially present in the conduction band or in defect sites. The number of photoelectrons is intrinsic to the material and pulse intensity, and is not subject to statistical fluctuation.
2. Conduction electrons, once formed, will be accelerated by the pulsed electric field. Their motion will become thermalized by electron-phonon collisions.
3. Heated electrons will, through impact ionization, release other electrons. The result is an exponentially growing "electron avalanche" leading to high concentration of free electrons and the formation of a plasma.

Because the pulse is short, the electronic energy increases much faster than it can be transferred from electrons to lattice. Therefore lattice thermal conductivity is unimportant until after the pulse is over.

4. Eventually, after the short pulse has ceased, the energy stored with the electrons is transferred to the solid as a whole. It then appears in visible forms as heat-related damage or (if the critical density of electrons was achieved) as plasma blowoff.

These observations are consistent with detailed calculations [4,5] which show that subpicosecond pulses allow insufficient time for a conventional electron avalanche -- one in which a very few seed electrons due to defects eventually collisionally ionize a large number of atoms. Instead, a large number of seed electrons are provided by the faster (at high intensities) multiphoton ionization. Thus, the energy deposition process is independent of defect concentration. The high power law dependence of multiphoton ionization rate on laser intensity explains the much sharper definition of damage threshold for shorter pulses. Finally, significant energy transfer from the electrons to the lattice does not occur during the ultrashort pulse. The actual damage occurs after laser energy deposition, i.e. after the pulse. In this sense, the physics of damage induced by ultrashort pulses is simpler than that of long pulses where a complicated balance between electron energy absorption and loss to various thermal processes causes sensitivity to details of structure and defect concentration.

Although the breakdown plasma is formed in a thin layer at the surface on the order of a wavelength thick, electron mean free paths are only on the order of  $10^{-7}$  cm. Since the plasma is many electron free paths thick, measured thresholds for surface damage are characteristic of bulk properties.

The fact that mechanical damage occurs well after energy deposition suggests independence of threshold determination from multiple pulse effects. The fact that the ablation rate per above threshold subpicosecond pulse is independent of the number of pulses [5] indicates that multiple pulse measurements merely serve to amplify the damage to easily observable levels.

## 7. THEORY OF SHORT PULSE DAMAGE

We have implemented these basic ideas into a consistent theory for short-pulse damage in homogeneous material. The theory consists of partial differential equations describing the changes in electron density and electron temperature during the pulse. The solutions to these equations reproduce the experimental results.

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  $U_I$  ( $h\nu \ll U_I$ ), the number density  $f(\epsilon, t)d\epsilon$  of electrons with kinetic energy between  $\epsilon$  and  $\epsilon+d\epsilon$  at time  $t$  is described by a Fokker-Planck equation,

$$\frac{\partial f(\epsilon, t)}{\partial t} + \frac{\partial}{\partial \epsilon} \left[ V(\epsilon, t) f(\epsilon, t) - D(\epsilon, t) \frac{\partial f(\epsilon, t)}{\partial \epsilon} \right] \equiv \frac{\partial f(\epsilon, t)}{\partial t} + \frac{\partial J(\epsilon, t)}{\partial \epsilon} = S(\epsilon, t) \quad (1)$$

where the Joule heating and loss to the lattice is

$$V(\epsilon, t) = \frac{1}{3} \sigma(\epsilon) E^2(t) - U_{phon} \gamma(\epsilon) \equiv R_J(\epsilon, t) - U_{phon} \gamma(\epsilon) \quad (2)$$

the energy diffusion coefficient is

$$D(\epsilon, t) = \frac{2}{3} \sigma(\epsilon) E(t)^2 \epsilon \quad (3)$$

and the conductivity per electron is

$$\sigma(\epsilon) = \frac{e^2 \tau_m(\epsilon)}{m^* (1 + \omega^2 \tau_m(\epsilon)^2)} \quad (4)$$

Here  $\epsilon$  is the electron energy,  $E(t)$  is the electric field oscillating at frequency  $\omega$ ,  $U_{phon}$  is the characteristic phonon energy,  $R_J$  accounts for Joule heating of electrons and  $\gamma(\epsilon)$  is the rate at which electron energy is transferred to the lattice. The quantity  $1/\tau_m(\epsilon)$  is the transport (momentum) scattering rate. Both  $\tau_m(\epsilon)$  and  $\gamma(\epsilon)$  are energy dependent. For energies in the conduction band of fused silica they vary by two orders of magnitude. The current  $J(\epsilon, t)$  represents direct heating and loss as well as an energy diffusion with coefficient  $D(\epsilon, \tau)$  which is proportional to both the conductivity and the laser intensity. The final term  $S(\epsilon, t)$  in Eq. (1) includes sources and sinks of electrons,

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

Impact ionization at rate  $R_{imp}$  was included assuming that excess kinetic energy is equally divided between the two resultant electrons,

$$R_{imp}(\epsilon, t) = -v_i(\epsilon) f(\epsilon) + 4v_i(2\epsilon + U_I) f(2\epsilon + U_I). \quad (6)$$

The source term  $S(\epsilon, t)$  also includes photoionization at rate  $R_{pi}(\epsilon, t)$ . The boundary conditions for Eq. (1) require the vanishing of the distribution at  $\epsilon \rightarrow \infty$  and the current at  $\epsilon=0$ . The important physical quantities  $n$  (electron number density) and  $\langle \epsilon \rangle$  (average kinetic energy per electron) are defined by the moments,

$$n(t) = \int_0^{\infty} f(\epsilon, t) d\epsilon, \quad n(t) \langle \epsilon(t) \rangle = \int_0^{\infty} \epsilon f(\epsilon, t) d\epsilon. \quad (7)$$

At high laser intensity, the energy absorbed from the laser field cannot be transferred to the lattice as fast as it is 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 remains fixed.

Numerical solution of the kinetic equation at constant laser intensity and without photoionization shows that an avalanche is established within a few femtoseconds for an intensity of  $1 \text{ TW/cm}^2$ . 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. Figure 1 shows, for a Gaussian pulse, the computed electron density that would result from multiphoton ionization alone and the density that occurs with the addition of impact ionization. As can be seen, seed electrons are produced by multiphoton ionization at the peak of the pulse, after which an avalanche produces a critical electron density.

Figure 2 compares the experimental values for damage threshold with the predictions of our theory.

## 8. DIELECTRIC DAMAGE

Laser induced damage to transparent optical elements is sensitive to surface properties and to the presence of structural imperfections that can enhance the electric field. It is important that all layers be prepared with care, to avoid introduction of damage sites. In our tests we are careful to select, by examination through a microscope, surface areas where visible irregularities are present. With this selectivity we obtain upper bound on the damage threshold for larger samples.

Our designs for multilayer dielectric gratings require materials with two contrasting refractive indices, termed high (H) and low (L). The requirement for high damage threshold eliminates many materials, and favors the use of metallic oxides as the high index layer. Silica has excellent qualities for the low index layer.

The following measurements indicate the range of damage threshold values occurring for oxides that we consider for use in dielectric gratings. These are the damage thresholds, in  $\text{J/cm}^2$ , for normal incidence 1053 nm radiation, 600 shots of 0.3 ps pulses with 500  $\mu\text{m}$  diameter spots

Dielectric:	Threshold, $\text{J/cm}^2$	Index
Tantala, $\text{Ta}_2\text{O}_5$	0.70	
Hafnia, $\text{HfO}_2$	0.83	1.9
Scandia, $\text{Sc}_2\text{O}_3$	1.23	1.8
Fused Silica, $\text{SiO}_2$	2.06	1.4

Because silica has a significantly higher damage threshold than do other oxides, it would be desirable to place it as the topmost layer, into which the grating is etched. Unfortunately the relatively low refractive index of silica requires that the grooves be much deeper than with higher index materials. Thus from manufacturing considerations it would be desirable, if possible, to use a high index material for the grating.

The values presented in the table are for normal incidence, whereas our gratings will be used at  $52^\circ$ . Simple arguments suggest that the damage threshold for incidence at angle  $\alpha$  on pristine surfaces should scale as the inverse of cosine  $\alpha$ . Our measurements on hafnia (see Figure 3) suggest that this scaling is reasonable.

## 8. MODIFICATIONS OF BASIC THEORY FOR STRUCTURED SOLIDS

The above picture refers to an infinite planar surface and a large laser spot size. In the theory, coupling of radiation energy into the material (and ultimately damage) occurs through the local electric field. It is the instantaneous local value of the electric field that drives damage.

In the multilayer grating, a complicated distribution of electric field occurs for an incident plane wave. The constructive interference that is responsible for the diffractive behavior of a grating or the reflective properties of a multilayer dielectric stack can enhance the electric field above values that would occur in unstructured homogeneous material. A more elaborate treatment is necessary to understand this situation.

In particular, local "hot spots" in the grating proper, or in the multi-layer, can increase the intrinsic susceptibility to damage. We have calculated the occurrence of such "hot spots" for practical grating designs, and have seen how their location is affected by design parameters. Although the "hot spots" due to constructive interference amplify the incident intensity, the effect may be somewhat ameliorated by the fact that the spots are small (sub wavelength) and thus electron diffusion and thermal conduction will begin to play a role. The inter-layer boundaries are prime sites for defects that can be easily damaged. Thus there may be an incentive to design multilayers with maxima away from these boundaries.

## 9. ELECTRIC FIELD DISTRIBUTION IN MLD GRATING

The distribution of electric fields around this structure shows a pattern of standing waves above the surface, as is to be expected from the combination of nearly equal amplitudes of incident and reflected light. Within the grating structure itself, the strength of the electric field exhibits a succession of nodes and of antinodes of diminishing magnitude. These are the result of the interference between upward and downward traveling waves.

There are two regions in which constructive interference is as large as the twofold enhancement of a mirror: at one corner of the (rectangular) grating, and at the first interface where a high-index material overlays a low index material. The high reflectivity of a quarter wave stack accompanies a standing wave pattern whose nodes align with the low-high index interfaces, and whose antinodes align with the high-low index interfaces. Figure 4 shows the electric field structure.

## 10. MULTILAYER GRATING DAMAGE

We have examined examples of multilayer dielectric gratings based on hafnia/silica layer pairs. The surfaces were observed to have many visible microscopic defects. On defect free areas we found a damage threshold (at the usage angle of  $52^\circ$ ) of  $0.76 \text{ J/cm}^2$ . The threshold on defects was somewhat smaller,  $0.65 \text{ J/cm}^2$ .

The morphology of the damage, as shown in Figure 5, is particularly striking. Our modeling predicts that there should occur regions of enhanced electric field strength along one edge of the ridges of dielectric gratings. Examination of SEM photographs of damage produced in multilayer gratings shows a consistent pattern of damage along and edge of the grating ridges. The damage appears first as a series of notches, roughly 0.1 micron apart. In regions of the grating exposed to relatively weak fields, the notches are short; in regions of high field the notches extend across the grating ridges.

## 11. CONCLUSIONS

We have measured the threshold for short pulse damage to dielectric material and to multilayer dielectric gratings. Our observations of damage morphology to multilayer dielectric gratings are consistent with the expected field enhancement along the edge of the grating structure.

## ACKNOWLEDGMENTS

This work was supported under the auspices of the U.S. Department of Energy at Lawrence Livermore National Laboratory under contract W-7405-Eng-48.

## REFERENCES

1. D. Strickland and G. Mourou, "Compression of amplified chirped optical pulses," *Opt. Comm.*, Vol. 56, pp. 219-221, 1985.
2. M. D. Perry and G. Mourou, "Terawatt to petawatt subpicosecond lasers," *Science*, Vol. 264, pp. 917-924, 1994.
3. M. D. Perry, R. D. Boyd, J. A. Britten, D. Decker, B. W. Shore, C. Shannon, E. Shults, and L. Li, "High-efficiency multilayer dielectric diffraction gratings," *Optics Lett.*, Vol. 20, pp. 940-2, 1995.
4. B. C. Stuart, S. Herman, and M. D. Perry, "Chirped-pulse amplification in Ti-sapphire beyond 1  $\mu\text{m}$ ," *IEEE J. Quant. Electron.*, Vol. 31, pp. 528-538, 1995.
5. 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.*, Vol. 74, pp. 2248-2251, 1995.
6. B. C. Stuart, M. D. Feit, S. Herman, A. M. Rubenchik, B. W. Shore, and M. D. Perry, "Nanosecond to femtosecond laser-induced breakdown in dielectrics," *Phys. Rev. B*, in press, 1995.
7. D. Du, X. Liu, G. Korn, J. Squier, and G. Mourou, "Laser-induced breakdown by impact ionization in SiO<sub>2</sub> with pulse-widths from 7 ns to 150 fs," *Appl. Phys. Lett.*, Vol. 64, pp. 3071-73, 1994.

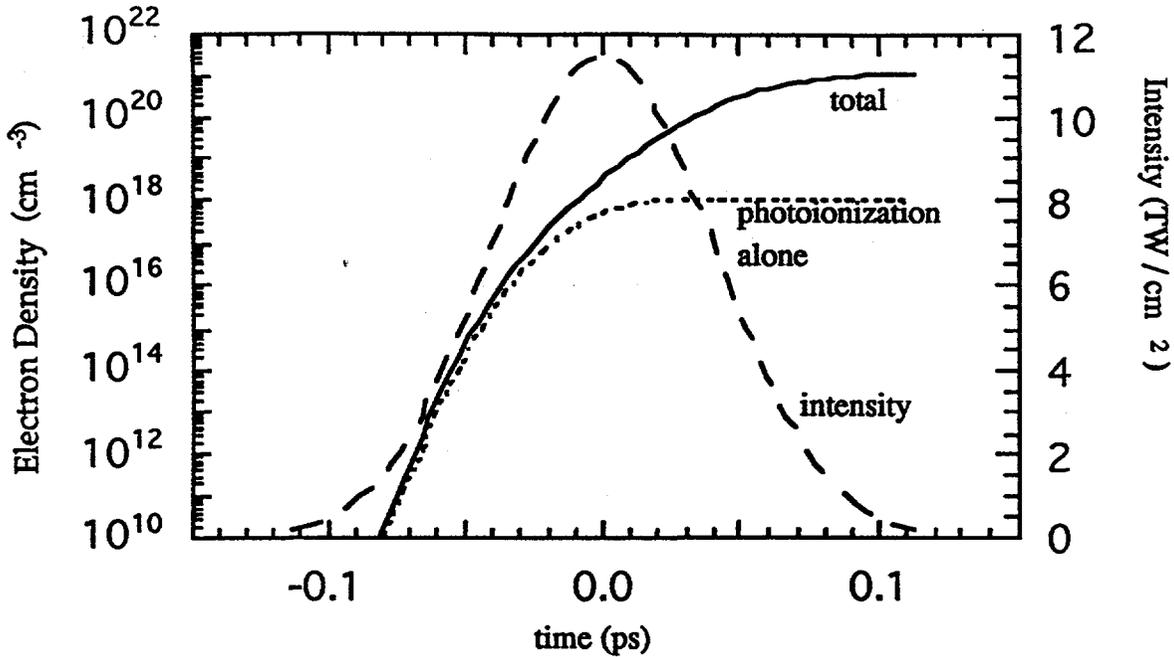
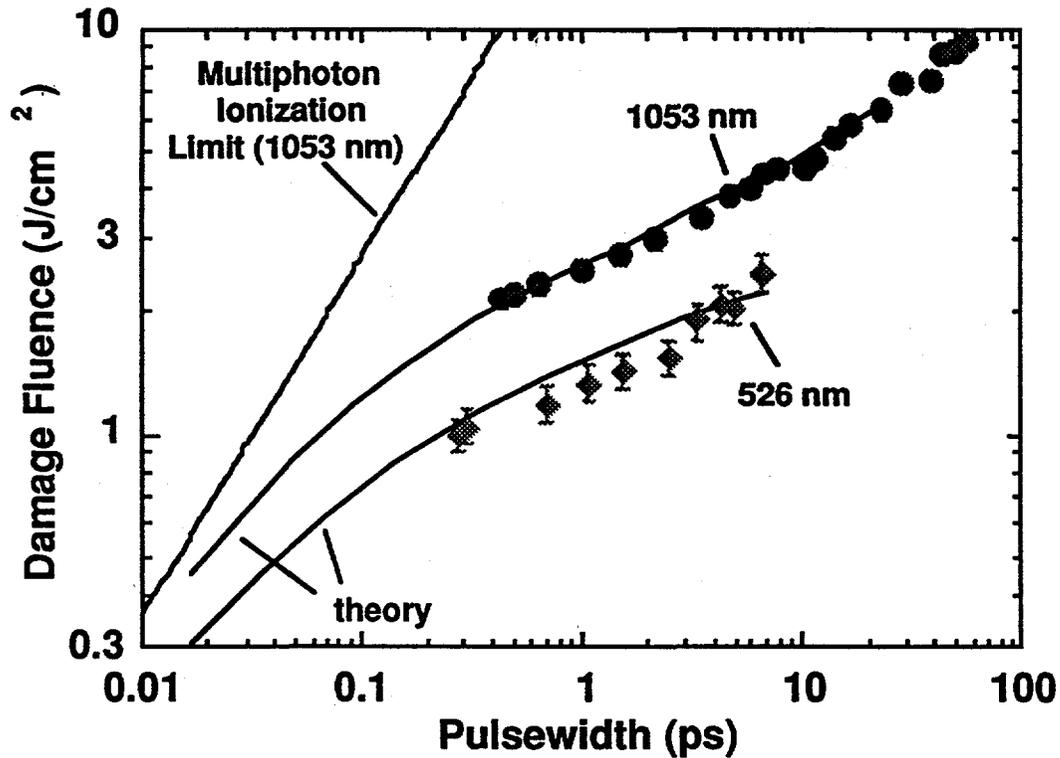


Figure 1. Total (solid line) and multiphoton produced (dotted line) are plotted along with the Gaussian pulse shape (dashed line).



vg-dallas04-14

Figure 2. Measured (dots) and calculated (solid lines) damage fluence for fused silica at 1053 and 526 nm. Dashed line indicates calculated damage limit due to multiphoton ionization alone.

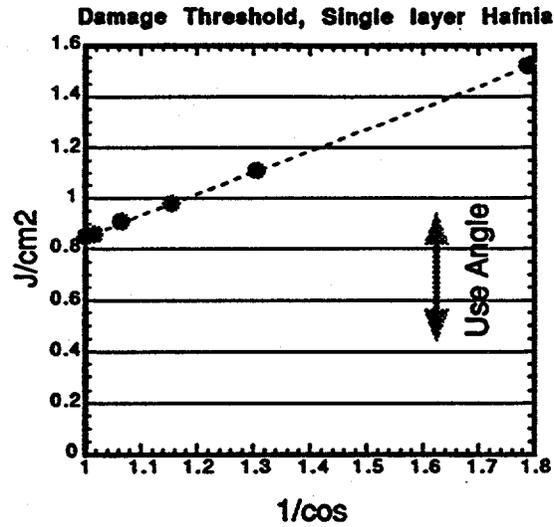


Figure 3. Damage threshold for 350 fs pulses on hafnia.

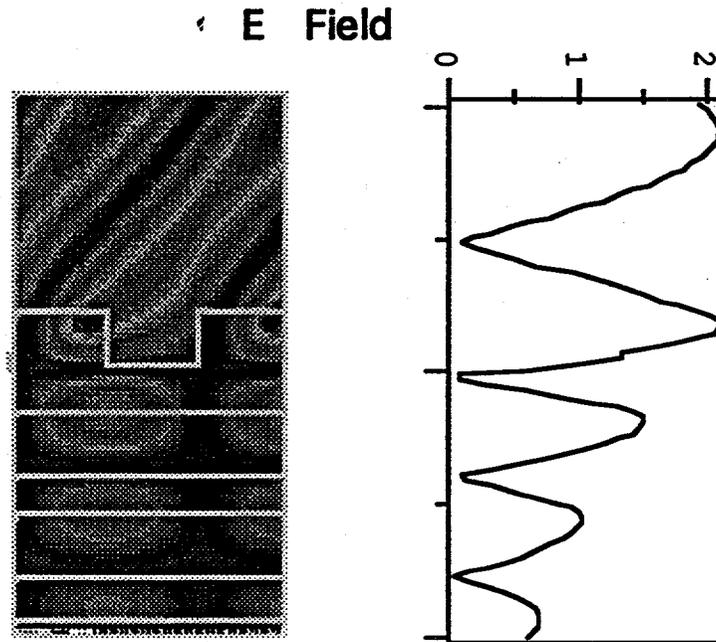


Figure 4. Electric field magnitude in a quarter wave HL stack surmounted by a lamellar etched grating. (Left): the refractive index vs. height above surface. (Center) contours of the electric field. An outline of the grating is superpsed. (Right). The electric field magnitude on a vertical slice through the regions of highest field.

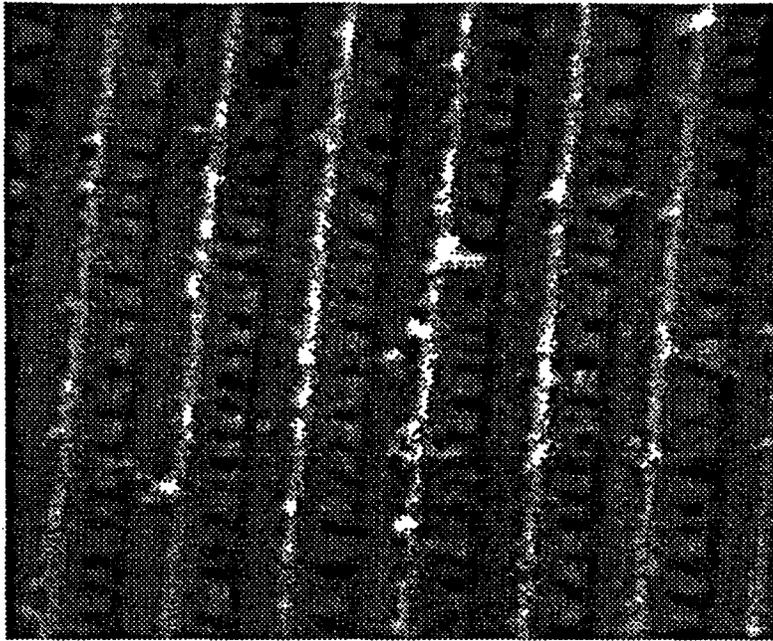


Figure 5. View of damaged grating surface, showing pattern of damage notches on (right-hand) edges of grating, coinciding with regions of enhanced electric field. The grating was a quarter wave HL stack, of H=hafnia and L=silica, surmounted by a lamellar grating etched into hafnia.