EXPERIMENTAL DETERMINATION OF DAMAGE THRESHOLD CHARACTERISTICS OF IR COMPATIBLE OPTICAL MATERIALS[*]

K. Soong[†], R.L. Byer, C. McGuinness, E. Peralta, Stanford University, Stanford, CA 94305, USA
E. Colby, SLAC National Accelerator Laboratory, Menlo Park, CA 94025, USA

Abstract

The accelerating gradient in a laser-driven dielectric accelerating structure is often limited by the laser damage threshold of the structure. For a given laser-driven dielectric accelerator design, we can maximize the accelerating gradient by choosing the best combination of the accelerator's constituent material and operating wavelength. We present here a model of the damage mechanism from ultrafast infrared pulses and compare that model with experimental measurements of the damage threshold of bulk silicon. Additionally, we present experimental measurements of a variety of candidate materials, thin films, and nanofabricated accelerating structures.

INTRODUCTION

Laser-driven dielectric accelerators have the potential to revolutionize the concept of particle accelerators. One major advantage of dielectric accelerators is their ability to generate accelerating gradients that are an order of magnitude larger than what is possible with current RF technology. The key to sustaining such extreme accelerating gradients is the relatively high damage resistance of dielectric materials. Compared to their metallic counterparts, dielectric materials can tolerate orders of magnitude higher electric fields before material breakdown occurs.

However as one might expect, there is a variation in the material breakdown limit even among just dielectric materials, and some consideration of the optimal dielectric is required. To further complicate the decision, the material breakdown limit is dependent on the properties of the drive source. In the case of laser-driven damage, the damage threshold of the material depends on the laser wavelength and the pulse duration. Therefore, when designing a dielectric accelerator, it is advantageous to consider a combination of dielectric material and drive wavelength that will result in the highest laser-damage threshold. In the case of the photonic crystal accelerator design [1], scaling the structure to the wavelength corresponding to the highest damage threshold will directly result in the highest accelerating gradient.

Unfortunately, there has been little prior work in developing an understanding for laser-induced damage in the regime where dielectric accelerators are most feasible, namely at mid-infrared wavelengths and with sub-picosecond laser pulses. This study aims specifically to provide some guidance for the optimal design of a dielectric accelerator, but the results presented are broadly applicable.

LASER-INDUCED DAMAGE MODEL

We present a plasma-based model for laser-induced damage. In this model, electrons in the dielectric medium are ionized to the conduction band through the absorption of photons from an incident laser pulse. The rate at which electrons are photo-ionized is described by the Keldysh model [2] with the laser pulse assumed to have uniform spatial intensity and a sech^2 temporal profile. Additionally, we account for changes in the density of conduction band electrons due to avalanche ionization and electron relaxation effects. We describe avalanche ionization with the Drude model [3], and model relaxation effects as a population decay. The total rate of change of the density of electrons in the conduction band is therefore described as:

$$\frac{d\rho}{dt} = W_P + W_A - \frac{\rho}{\tau}, \qquad (1)$$

where ρ is the conduction band electron density, W_P is the photoionization rate, W_A is the avalanche ionization rate, and τ is the relaxation time constant.

If electrons in the conduction band reach a critical density, then damage will occur in the form of material ablation. This critical density, n_e, is defined as: the point at which the plasma frequency and the laser frequency are resonant. More concisely, when

$$\omega_l = \omega_p = \sqrt{\frac{4\pi n_e e^2}{m^*}}, \qquad (2)$$

where ω_l is the laser frequency, ω_p is the plasma frequency, e is the electron charge, and m^* is the effective mass of the electron.

The laser-damage threshold is correspondingly defined as the minimum laser fluence required to reach the critical electron density in a single pulse. The calculated laser-damage threshold of bulk silicon by a 1ps laser pulse is shown in Fig. 3.

EXPERIMENTAL SETUP

To experimentally determine the laser-damage threshold of candidate materials, we designed the pump-probe experiment diagrammed in Fig. 1. An optical parametric amplifier (OPA) served as our tunable laser source, providing sub-picosecond duration pulses in a wavelength range from 1μm to 3μm and at a repetition rate of 600Hz. The

pulse duration was measured using an intensity autocorrelator and the spectral purity of the OPA was verified using a commercial spectrometer.

The IR pulses from the OPA were focused with a $f/1$ lens onto the surface of the test sample, which was held in place by an aluminum mount. Mounted on the same plane as the test sample was a set of knife edges used to determine the laser spot size. Upstream of the interaction point, a pellicle beamsplitter in conjunction with a pyroelectric detector was used to sample each pulse. The laser pulse energy was adjusted with a motorized variable neutral density (ND) attenuator, placed upstream of the beam splitter. Prior to each set of measurements, the pyroelectric detector was calibrated against an energymeter placed at the interaction point.

In order to detect damage in situ, we reflected a low power CW HeNe laser off the surface of the test sample, taking care to co-align the reflection point with the interaction point. Downstream of the interaction point, the intensity of the reflected HeNe laser was measured with a silicon photodetector. Damage to the test sample will manifest as a distortion to the sample's surface, causing a decrease in the measured HeNe intensity.
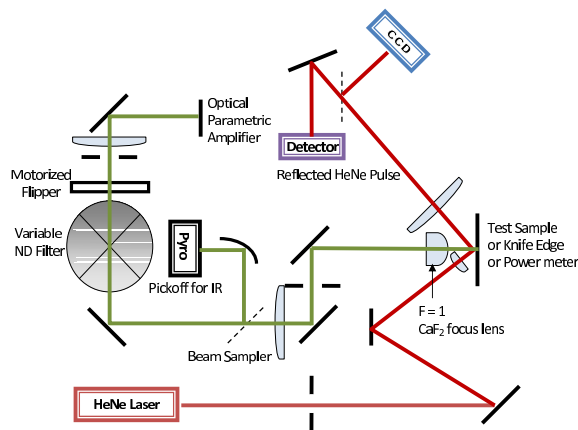
Figure 1: A diagram of the pump-probe experiment used to determine laser-damage threshold. The intensity autocorrelator and the spectrometer are not shown.

Each data set consisted of one-hundred and eighty test sites on a single test sample, with each test site separated by a distance of 350μm. For each test site, we impinged 36,000 pulses, recording the average energy per pulse and whether or not damage occurred. If damage did not occur, the pulse energy was increased using the variable ND attenuator before moving to the next test site, and vice versa. In this method, the pulse energy was converged to a value corresponding to the material's damage threshold. Damage threshold values reported in this paper are the converged values.

RESULTS

Silicon

Silicon has become ubiquitous in the nanofabrication industry and with the overwhelming knowledge-base for processing silicon, it becomes one obvious candidate material. We measured the laser-damage threshold of bulk silicon, in the form of a polished silicon wafer, for a span of wavelengths in the near-to-mid infrared. The laser pulse width was approximately 1ps across all measurements and the laser spot had a Gaussian FWHM on the order of 10μm. An SEM image of a typical damaged test site is shown in Fig. 2. The fringe patterns present in the image is typical of multi-shot laser-damage. A direct comparison of the laser-damage model and experimental data for bulk silicon is shown in Fig. 3.

Figure 2: An SEM image of a typical test site after laser induced damage. The fringe pattern is characteristic of multi-shot laser damage.
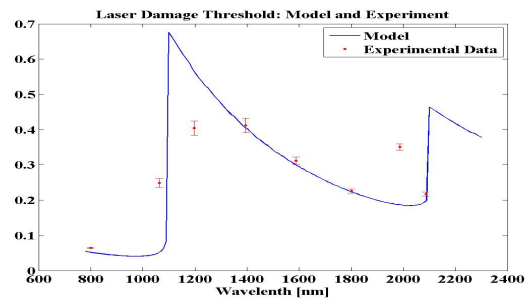

Figure 3: Laser-damage threshold of silicon as predicted by our model (blue) and as measured (red). The wavelength range includes (partially) one, two, and (partially) three-photon ionization regimes.

The laser-damage model indicates that the damage threshold of silicon does not follow a monotonic pattern. The predicted discontinuities in the model correspond with a transition from n to n+1 photon ionization; the location of these discontinuities is determined by the material's bandgap. As can be expected, the significantly smaller absorption cross-section for many-photon processes results in a lower density of conduction band electrons, or correspondingly a higher damage threshold when transitioning from n to n+1 photon ionization. However, when comparing predicted damage thresholds within the same n-photon

ionization band, we see that there is a gradual decrease in damage threshold with increasing wavelength. This gradual decline in damage threshold is explained by the lower critical electron density, indicated in Eq. 2. These physical effects are not unique to silicon, and we expect to see these features for most dielectric materials.

We find reasonable agreement when comparing our experimental data with the laser-damage model. The model accurately predicted the damage threshold at wavelengths sufficiently distant from the material bandgap. Near the bandgap however, rather than the sharp discontinuities predicted by the model, our results indicated a smoother transition. It is unlikely that this effect was caused by a broad laser spectrum, since the spectral purity was verified prior to each measurement and the FWHM line width was confirmed to be less than 20nm. More likely, this effect is due to an impurity on the surface of the test sample. Although the test samples were cleaned with Isopropyl alcohol prior to each measurement, the measurements were performed in air with no additional care taken to control for dust and debris. We would expect local field enhancement from particles on the sample surface, therefore decreasing the damage threshold, particularly in the case of very high fields.

Other Materials

It is not always possible to design an accelerating structure to operate at the wavelength for maximum laser-damage resistance. An accelerating structure requires sub-wavelength features, which imposes a limitation on the minimum laser wavelength; this limitation is set by the fabrication technology. On the other extreme, the presence of material absorption bands at mid-infrared and long-infrared wavelengths imposes a limitation on the maximum laser wavelength. It is therefore of interest to find an optical material that has a high damage threshold at a wavelength that is compatible with both current fabrication and laser technology. With this motivation, we present damage threshold measurements of a variety of materials at a wavelength of 800nm; this data is compiled in Table 1. Fabrication at this wavelength is easily accomplished with 266nm lithography techniques, and Ti:Sapphire high power lasers are commercially available.

Table 1: Damage Threshold Measurements at λ=800nm

Material	Thickness	Bandgap	F_{th}[J/cm^2]
Al$_2$O$_3$	1000μm	9.9eV	4.90$\pm$0.29
SiO$_2$ (Quartz)	1000μm	8.9eV	4.10$\pm$0.50
ZrO$_2$/Y$_2$O$_3$	15nm	5-7eV	3.97$\pm$0.16
HfO$_2$	<200nm	5.8eV	3.63$\pm$0.36
Si$_3$N$_4$	100nm	5.1eV	0.65$\pm$.05
Si	1000μm	1.1eV	0.14$\pm$.02

The damage threshold data shows a strong positive correlation between the damage threshold, F_{th}, and the material bandgap. Although thin films have additional physical mechanisms contributing to the laser-damage, they were not considered in this comparison.

Accelerating Structures

The laser-damage threshold measurement of bulk materials give a good sense of the performance of an accelerator structure, however it is more realistically an upper performance limit. An accelerator structure will necessarily contain features which will contribute to local field enhancements, thus diminishing the structure's damage tolerance. For the proposed grating structure [4], numerical calculations with the code HFSS reveals that the local field enhancement effect instigates laser-induced damage at a fluence equal to 0.45 times that of the bulk material's damage threshold. We verify this factor with experimental measurements of a fabricated grating structure and find good agreement, as shown in Table 2. In each case the laser wavelength was matched to the grating period, to conform with the simulation. These measurements, along with simulations, indicate that the grating structure can achieve an accelerating gradient in excess of 300 MV/m.

Table 2: Damage threshold measurements of a grating-based dielectric accelerator, compared to bulk material.

	Silica Grating	Bulk Silica	Ratio
λ= 800nm	1.54$\pm$0.07 J/cm^2	3.23$\pm$0.11 J/cm^2	0.48
λ=1500nm	1.85$\pm$0.11 J/cm^2	3.46$\pm$0.14 J/cm^2	0.53

CONCLUSION

We have described in this paper a model for the laser induced damage mechanism of dielectric materials. To verify the model, we experimentally determined the damage threshold of silicon over a range of wavelengths and found that the model accurately predicted the damage threshold. Additionally, we explored the damage threshold properties of six different materials and found a correlation between the material's bandgap and its damage threshold. Lastly, we measured the damage threshold of a grating accelerator structure and found good agreement with simulation. Future work will continue the testing of high damage-resistant optical materials, as well as accelerator components. In addition, techniques for improving damage resistance, such as film coatings, hydrogen annealing, and laser conditioning, will be explored.

ACKNOWLEDGMENT

REFERENCES

[1] B. Cowan, "Three-Dimensional Photonic Crystal Laser-Driven Accelerator Structures," SLAC-PUB-12090 (2006).

[2] L. V. Keldysh, "Ionization in the Field of a Strong Electromagnetic Wave," Soviet Physics JETP, Vol. 20 (1965) 1307.

[3] B. C. Stuart, et al., Phys. Rev. B 53 (1996) 1749.

[4] E. Peralta, et al., "Fabrication and Measurement of Silica Grating Accelerator Structures," PAC 11, MOP096 (2011).