40 year retrospective of fundamental mechanisms

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Retrospective

Abstract: Fundamental mechanisms of laser induced damage (LID) have been one of the most controversial topics during the forty years of the Boulder Damage Symposium (Ref. 1.) LID is fundamentally a very nonlinear process and sensitive to a variety of parameters including wavelength, pulse width, spot size, focal conditions, material band gap, thermal-mechanical prosperities, and component design considerations. The complex interplay of many of these parameters and sample to sample materials variations combine to make detailed, first principle, models very problematic at best. The phenomenon of self –focusing, the multi spatial and temporal mode structure of most lasers, and the fact that samples are 'consumed' in testing complicate experiential results. This paper presents a retrospective of the work presented at this meeting.

Key Words: Fundamental Mechanisms, Laser-Induced Damage (LID), Laser-Induced Breakdown (LIB), Self-Focusing, Pulse-width Dependence, Wavelength Dependence, Band-Gap Dependence. Surface Ripples, Sol Gel Coatings, Scaling Laws.

Introduction:

I accepted the invitation of Vitally Gruetev and the co-Chairs of our meeting to give the invited talk on fundamental mechanisms at the 40th anniversary Boulder meeting with the full intention of reviewing the literature of this meeting and elsewhere on this topic and giving a proper review, with appropriate attribution to progress made and a thoughtful summary as to progress made and challenges remaining. I agreed to do this to try to transition back into active science from my current life as a university bureaucrat.

However, my "day job" has proved all consuming, leaving little time and even less energy to devote to this task. The net result is a "Retrospective" on the topic rather than the thorough review I had hoped to present.

Last year we marked the passing of Art Guenther. I ask that we keep alive his memory by having an intense, professional, and fun debate about LID at this the 40th Anniversary Boulder Damage Symposium, the meeting that he created for us. Please remember also Art's classmate and our friend and colleague, Jean Bennett, who passed away a few months ago.

Nonlinear Processes:

LID is a very non-linear process. LID in transparent dielectrics is the most complicated since it depends on:

- materials prosperities (most of which are not properly understood);

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- laser parameters (wavelength and pulse width); and
- the exact conditions of the experiments (mode structure and focusing conditions).

And we learned most of this from the first paper at this meeting on this topic by Bliss! (Ref. 2)

There has been much great work in multi-phonon absorption new materials new methods, and design consideration which have come from this meeting. In addition, this progress was enabled by precision measurements of absorption, scattering, and other critical parameters. (Ref.1)

Among the many important fundamental results reported here are multi-phonon limits of linear absorption, very good models for LID to metals, the role of stress in high power CW applications, the effects of absorption on beam quality, among others. Many of these advances were made possible by progress in precision measurement of small levels of absorption, surface roughness, and scattering.

Linear absorption by defects, inclusions, and contamination remains a practical concern, but the fundamentals are reasonably well understood.

Laser Induced Breakdown (LIB), the catastrophic, threshold-like process in highly transparent materials, is where the real controversies have been and where understanding still lags. The quest for understanding fundamental limits runs head on with the many varied and complex non-linear processes involved. The problem has been further complicated by what I believe were some fundamental errors early in this quest for understanding.

The subject of fundamental limits on LID has produced wide agreement and extreme disagreement. Almost everyone will agree that for damage to occur there must be absorption of laser radiation by the material. However, beyond that, there is little agreement, or perhaps more properly stated, no detailed understanding as to how a material goes from highly transparent to totally absorbing within just a few pico seconds. (Ref. 3) The discussion of LIB mechanisms is a discussion of the process or processes by which this sort of "change of phase" occurs.

One reason to seek such understanding is to be able to do reasonable scaling with respect to various parameters such as pulse width, wavelength, materials prosperities, beam prosperities, etc. In addition, it is thought that if indeed the fundamental mechanisms of failure are understood, ways to improve materials could be found for materials not yet at the fundamental performance limits.

Self-Focusing:

A major issue in the design of laser systems has been self-focusing (Ref. 4), particularly in gain media. This was the subject of many excellent theoretical and experimental papers in the early days of the Boulder damage conference. In the first 30 years of this meeting, there were 48 papers and 614 references on this subject (Ref. 1).

Self-focusing is not in itself a mechanism for LIB; rather, it is a mechanism for concentrating the beam so that local intensities reach critical values at which other nonlinear process occur. Among the triumphs was the understanding of how to pick and sometimes design materials to minimize the nonlinear refractive index, n2, which produces self focusing.

A very practical advance was the understanding that self-focusing can be managed by careful control of the mode of laser oscillators and properly accounting for nonlinear propagation of beams in large oscillator-amplifier systems. The key is to avoid and/or remove high spatial frequencies from the beam. In practical terms, this means over sizing the optics so that the beam is not clipped and the use of spatial filters to remove any high spatial frequencies, which may occur during beam propagation. In this way, self-focusing in gain media is effectively managed in large laser systems such as those used in laser fusion experiments.

However, the problem often reappears as laser makers try to maximize energy output from gain media of limited size by multimode operation, a problem made worse by trying to compact multimode systems in limited space. This prevents the loss of high spatial frequencies and, in turn, results in catastrophic damage due to small scale self-focusing.

The influence of self-focusing in damage experiments is a bit more problematic and has been a major issue in understanding the parametric dependence of LIB in transparent solids. Examples include the fact that the critical power for self-focusing scales as the square of the wavelength, as the inverse of the pulse width, multiple mechanisms exist for self-focusing and these can scale as the spot size and pulse width together (electrocution), and self-focusing can exhibit high dispersion. Thus, self-focusing can be confused with wavelength, pulse width, and spot size dependence of LIB.

Early Work in LIB:

Early observations of LIB include the following:

- Abrupt truncation of the beam transmitted through the sample, i.e., threshold like behavior;
- bright, hot plasma accompanying damage; and,
- Damage at the peak of the pulse when at threshold irradiance.

These observations, plus the lack of any reasonable alternative for energy coupling in otherwise transparent materials with band gaps 5 to 20 times the photon energy of the laser that produced damage, led early investigators to conclude that LID was due to electron avalanche breakdown.

Early controversies and discrepancies were due to issues in mode structure of lasers used in measurements and confusion in quoting rms vs peak E-fields in breakdown measurements.

However, the real problems are (1) trying to describe such a nonlinear process in enough detail that materials may be improved, and (2) that dependence on pulse width, wavelength, and spot size can be made with confidence.

This situation was made more complex by the simple fact that no precursor of LIB was available in early experiments (and remains elusive), and experiments are hard to replicate, since the samples are consumed in the experiment and new samples often produce different results.

In addition, there was the key question of where the initial electrons came from; that is, is LIB an intrinsic or extrinsic phenomenon?

I want to now use a bit of hindsight to look back at some work that escaped proper attention by our community.

Early work (1968) in Japan by Yasojima (ref. 5) showed laser damage in NaCl at 10.6 microns showed LIB, i.e., avalanche like, behavior. The author speculated that this avalanche was seeded by non-linear processes and the author demonstrated that the threshold of LIB could be substantially reduced by "seeding" the damage with a flash of blue light, thus producing "starter" electrons for the LIB.

This was a remarkable bit of work and went unnoticed in work reported at the Boulder Damage Symposium and in other Western and Soviet literature on this subject. I first became aware of this work while doing a thorough search of literature in the preparation of my PhD thesis in 1978.

Based upon his seeding experiments, Yasojima speculated that LIB was initiated by either multi-photon absorption or tunneling to produce free carrier that could be rapidly accelerated by the several mega volt E-fields associated with focused pulsed laser beams.

The work presented in the first 40 years of Boulder meetings mostly produced further evidence for the speculation of this unrecognized scholar!

<u>LIB in Gasses.</u>

There was much work in LIB in gasses in the mid-1960s (Ref. 6), which mimicked what was seen in solids. First there was the 'clean' vs. 'dirty' gas results. Specifically, air breakdown seemed to show little frequency dependence and indeterminate pulse width and spot size dependence. However, this confusion was greatly reduced by using very pure gasses, free from any particulates that could initiate LIB.

Theories of avalanche breakdown were thoroughly tested by reducing the pressure of the gasses to the point where momentum transferring collisions could not occur at rates needed to allow an avalanche to develop.

Finally, controlled experiments with clean gasses, in low pressure, clearly showed multi-photon absorption (nPA) production of electrons consistent with theory.

In essence, these workers had well defined, re-usable and repeatable samples and controlled critical parameters needed to test avalanche models and measure field dependent ionization rates, parameters that still elude us in LIB in solids. Careful attention to the gas breakdown work would have kept authors out of the trap of assuming that they were seeing intrinsic LIB in solids.

Note that we had some strong indications of differences between air breakdown and LIB in solids. One can do surface damage experiments at one micron without worry about LIB in the air. However, LIB in air prevents damage on optical surfaces at 10.6 microns, mandating that experiments be done in a vacuum. Thus, laser damage experimenters and theoreticians should have noted the vast difference between LIB in gasses and solids, e.g., much stronger dependence in air then for solid materials.

<u>Entrance vs. Exit Surface Damage: A Fundamental Mechanism Understood.</u>

One of the more interesting LID phenomena is the simple fact that for an uncoated dielectric an incident parallel beam damages the rear or exit surface of a sample at a lower input flux then the front or entrance surface, even though less energy is incident on the rear surface due to Fresnel losses at the iterance surface. This initially surprising result led to some really interesting, complex explanations by some very notable scientists in the published literature, all of which turned out to be dead wrong.

The explanation of this effect (Ref. 7) is elegantly simple and gives precise, repeatable LID results. The damage threshold is lower for the rear surface because the E-field associated with the beam is larger at the rear surface even though the incident energy is less than that on the front surface.

This is understandable from simple boundary conditions for electric fields from Maxwell's equations. The field at the front surface is less because the field from Fresnel reflection is out of phase with the incident field, whereas the reflected field at the rear surface is IN PHASE with the incident field. Thus the field on the exit surface is larger and the exit surface damages before the entrance surface.

Once understood this effect is so obvious as to be a homework problem for an undergraduate class in optics. And it is easily tested and yields the exact results expected.

This simple result led to new ideas in designing thin film coatings and in general "managing" the E-field of a laser beam in an optical train to minimize damage. This result is discussed in detailed by Brian Newman in his review of thin film damage in this volume.

The understanding of the field effects in this simple experiment led to speculation by N. Bloembergen (Ref. 8) that surface damage was lower then bulk because of field enhancement at defects. The understanding of this field dependence of surface damage also led to greater confidence in the avalanche theory and other field dependent models.

However, there are still a couple of field effects regarding surface damage, which remain a puzzle, and are discussed later in this paper, i.e., lack of larger field enhancement in specific defects and lack of surface ripples in laser damage sites on the entrance surface of dielectrics, particularly at 10.6 microns.

Intrinsic or Extrinsic Damage?

The Harvard group, led by Nobel Laureate Nicolas Bloembergen (Ref. 9) and co-workers, published a very impactful and often referenced papers that claimed that LIB was an intrinsic prosperity of materials. This claim was based up on two factors:

1. The observations of breakdown-like behavior, which I previously mentioned, and,
2. The observed frequency dependence (really very little frequency dependence) of LIB in NaCl from 0.35 to 10.6 microns.

The Harvard group assumed that the breakdown irradiance was an intrinsic property of a material; thus, any sample to sample variations measured by others were due to poor quality samples and any spot size dependence was purely due to self-focusing.

Other work produced different results (Ref. 10, 11, 12, 13) than the Harvard group reported including:

1. There are just not enough free carriers present in good insulators (highly transparent materials) to initiate damage for very small spot sizes, i.e., it takes at least a few free electrons to be present to initiate the avalanche. (Ref. 10)

2. Better materials with higher than the so call 'intrinsic' thresholds became available, some with a factor of 20 higher LIB thresholds than the previously reported 'intrinsic' results.

3. Subsequent frequency dependence measurements showed a decrease in threshold at 0.5 microns, inconsistent with a pure avalanche model.

4. Evidence was produced, which indicated that not all spot size dependence was due to self- focusing. Lack of polarization dependence of LIB (as would be expected if self-focusing was initiating damage) was used this to rule out self-focusing in some spot size dependence measurements.

Note that the increase in LIB threshold measured by the Harvard group up to 0.3 microns was probably due to the fact that as one moves to shorter wavelengths, diffraction limited spot sizes get smaller, and spot size dependence could be interpreted as wavelength dependence.

Current Understanding.

Where does this leave us? Figure 1 taken from Ref. 14 (figures 7 and 8 in Ref. 14) is an illustration of the problem. Here the spot size is constant and the pulse width and wavelength dependence is given for NaCl and SiO2. Note that in this case the spot size was the same at both wavelengths, and lack of self-focusing was confirmed from measurement of lack of polarization dependence in the LIB thresholds.

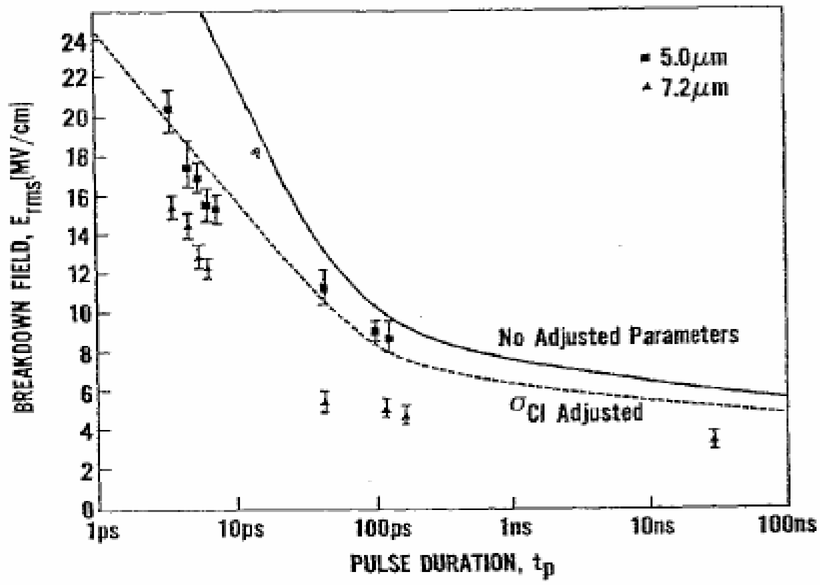


Figure 7. The RMS breakdown field data for NaCl (78-NC-6) at 1.06 μm are plotted as a function of pulse duration, t_p. The solid line and dotted line were obtained from the theory developed by Sparks et al. [28] for NaCl at room temperature. The dotted line uses a different value for the absorption cross section for Cl ions in the theory than the solid line.

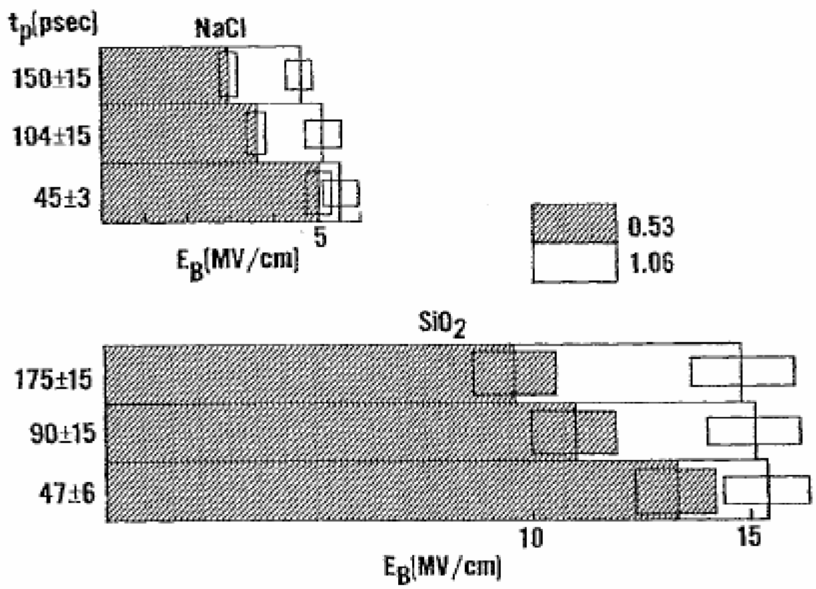


Figure 8. Wavelength dependence of the breakdown field E_B for NaCl and SiO_2 for a variety of laser pulsewidths. All the above data was taken on the same sample of NaCl and the same sample of SiO_2. The 1.06 μm thresholds are taken from Ref. 2 and are interpolated from measurements made at spot size 6.1 and 10.3 μm.

Figure 1. The two figures (7 and 8) above are taken from Ref. 14. Ref. 28 in the caption is reference 15 in this paper.

The decrease in LIB field with pulse width is consistent with an avalanche process for these pulses in the pico second regime. However, the wavelength dependence is not consistent with a purely avalanche model, and not strong enough to suggest a multi-photon model for LIB. Note that this model is very much dependent on the very nonlinear ionization rates, which, in fact, are estimated from DC measurements.

These results are not inconsistent with a model, which assumes an avalanche initiated by multi-photon or by tunneling.

After 40 years "not inconsistent" is about the best one can do. Quantitative information about avalanche ionization dependence of laser fields in solids is not available; accurate parameters for nPA or tunneling models; and definitive information indicating that all critical parameters has been considered is not available.

Measurement of Critical Parameters Leading to LIB.

Part of the problem in sorting out fundamental mechanisms of damage is that critical parameters are not easily measured, or in some cases simply estimated. For example, the avalanche ionization rates used in figure 1 are from DC measurements. It is unlikely that such rates can be extrapolated to 10x14 Hertz with sufficient accuracy to definitively test theoretical models.

New techniques and shorter pulses allow more quantitative measurements of nonlinear parameters, e.g., Z-Scan (Ref. 16) for n2, sign of n2, nPA, excited state absorption, etc...

Figure 2 is an example of the Z-scan technique for measuring nonlinear refraction -- with sign as well as magnitude -- and nonlinear absorption (reference ZnSe paper.) This powerful technique reduces what used to be a PhD dissertation problem to a few minutes of simple measurements. In addition this measurement gives the sign as will as the magnitude of n2. Note that for ZnSe the sign of nonlinear refraction changes from one to one-half micron. Measurements at different pulse widths are used to separate out various nonlinear mechanisms, e.g., electrostriction and electronic nonlinear refractive indices. By fully opening the aperture in the front of the detector, one can measure nonlinear absorption and pulse width dependence can separate excited state absorption from nPA.

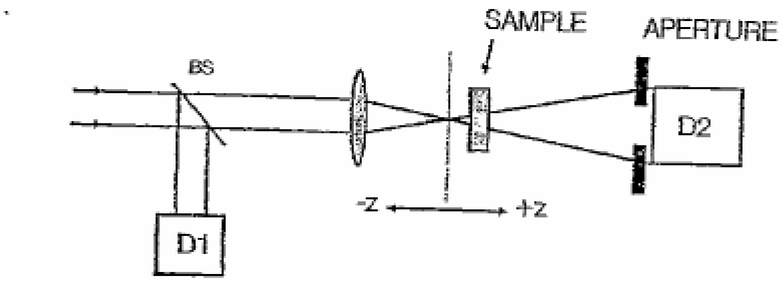


Figure 2. Z-Scan Technique from Ref. 16 figure 4. Note that the sample is scanned through the beam focus. The ratio of the readings of D1 and D2 is measured. The fit from the curve of this ratio vs. Z (position relative to the focus) give the nonlinear index of refraction. If the aperture (D2) is fully open and captures all the transmitted beam then nonlinear absorption is measured.

Note that ZnSe is a 2PA material at 0.5 microns and a 4PA material at one micron. The nonlinearity shown is for pico second pulses and is due to electronic effects. For nano second pulses, electrocution can also play a role for small spot sizes.

Figure 3. Measurement of n2 (solid circles) at 1.06 microns and 0.53 microns (open circles) showing the sign change for ZnSe (positive n2 and negative n2 respectively.) This data is figure 8 in reference 16.

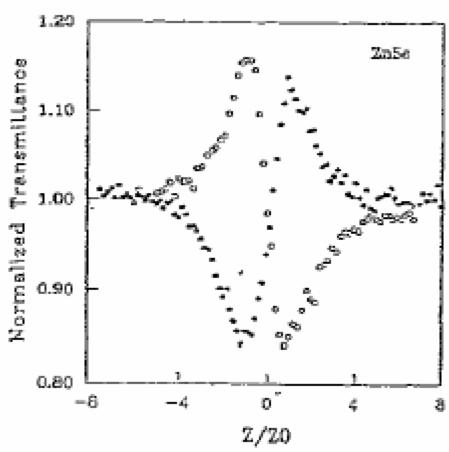
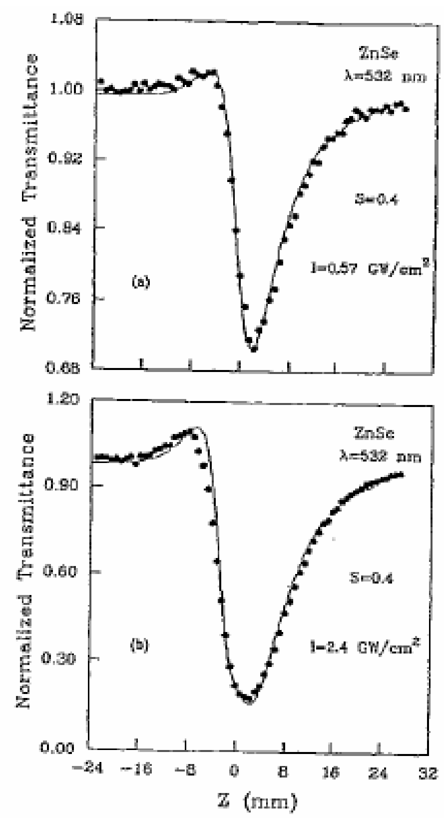


Figure 4. This is figure 6 from ref. 16 and it shows the power of the Z-Scan technique to help understand relevant nonlinear interactions, including 2PA, excited state absorption (free carrier absorption), and nonlinear refraction for ZnSe at 0.53 microns and 30 psec pulses. The bottom curve shows the results of increasing the irradiance by about a factor of 5, and the fit is excellent indicating that the essential physics is accounted for in this measurement.



Complete understanding of fundamental mechanisms will await such detail, reproducible, non-destructive measurements of processes leading to damage.

<u>The Role of nPA.</u>

What about nPA? Can this process be the mechanism for LIB? Reference (Ref. 17) attempted to measure the onset of LIB through measurements of samples of different band gaps at two laser frequencies. Measurements were made for 2, 3, and 4PA materials at one micron for 49 psec pulses.

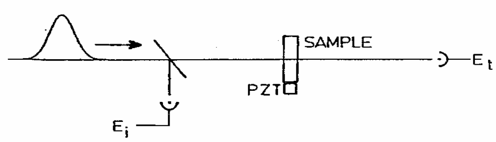
This result showed that 2 or 3 PA carriers generated prevent damage by defocusing the beam.

What happens at 4 PA? The results are strikingly different. For 4PA carrier generation rate is not sufficient for free carrier de-focusing to prevent damage but sufficient, and laser electric fields high enough, to seed an electron avalanche process.

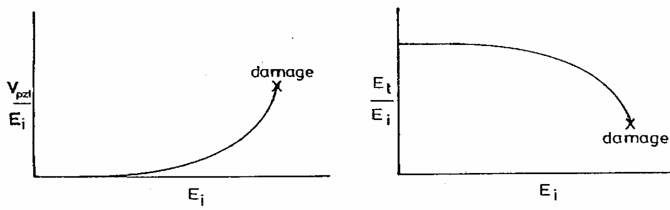
Figure 5. The figure below shows the effects summarized above. The data are for 40 psec pulses at 1.06 microns. These data are from Ref. 17.

NONLINEAR SPECTROSCOPY

NONLINEAR ABSORPTION IN II-VI MATERIALS



Transmission and Photoacoustic Monitor Nonlinear Absorption



PAS Signal vs. Incident Energy

Nonlinear Transmission vs. Incident Energy.

Material	Dominant Absorption	Damage Threshold (GW/CM^2)
CdTe	2 photon + free carrier	2.0 (front surface)
ZnTe	2 photon	0.46 (front surface)
CdS	3 photon + free carrier	19.0 (front surface)
CVD ZnSe	3 photon + free carrier	12.0 (front surface)
CVD ZnS	4 photon	15.1 (exit surface)
Cleartran (ZnS)	4 photon	18.1 (exit surface)

Summary of nonlinear absorption and damage threshold measurements in the II-VI materials. Data shown are for 40 psec pulses at 1.06 μm.

Progress in using femto second pulses, interference techniques (see for example the paper by Serge Garnov in this volume) (Ref. 18) form measuring carrier generation at focus, etc. offer hope of helping understand LIB in highly transparent materials.

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Remaining Mysteries Related to Surface Damage.

Many experiments have shown surface ripples inside the laser damage area. These structures have been shown to be related to pre-existing surface defects, e.g., scratches.

Sipes and Van Dreill (Ref. 19) and Temple and Soileau (Ref. 20), and many other workers explored the theoretical explanation of surface ripples with accompany LID of surfaces. Several models explain the orientation (orthogonal to the polarization direction the laser radiation) and the spacing (varying from the wavelength of the laser light in vacuum to the wavelength in the medium.)

At 10.6 microns (ref. 21) these structures are omni present for damage to the exit surface of NaCl and other dielectrics. Such structures have never been observed in LID sites on the entrance surface of dielectrics. Published models for these phenomena are symmetric with respect to the entrance and exit surfaces.

Finally, the huge field effects due to surface defects are just not observed. Various models predict factors of 10 to 100 enhancements at surfaces due to certain types of defects. Given the highly nonlinear nature of LIB, one would expect to see orders of magnitude variations in surface damage due to such resonant defects. No such variation is seen.

The most damage resistant surfaces reported are porous surfaces of sol gel coatings (ref. 21) These surfaces consist of highly disordered, high density voids. Field enhancement theory would seem to predict poor performance of such surfaces. Just the opposite is observed! Certainly more work is needed on this subject.

Scaling Laws:

Even after more than 40 years of study, the parametric dependence of the various mechanisms of LID are not well enough understood to allow accurate scaling models that can ensure adequate design of laser systems (see for example ref. 22.)

However, there is a "rule of thumb," which gives a good starting point in estimating limits imposed by LID:

$$E_d = (10\ \mathrm{J/cm^2})(t_p/1\ \mathrm{nsec})^{1/2} \qquad \text{(eq. 1)}$$

where E_d = damage threshold fluence in Joules /cm^2

t_p = laser pulsewidth in seconds.

Eq 1 gives an *approximation* of the threshold fluence to within plus or minus an order of magnitude for optical materials (transparent dielectrics) over the wavelength range from the UV to the infrared. This is admittedly a gross over simplification. The proper use of this equation is simply to get an idea as to whether or not one should be concerned about the possibility of LID. Laser systems designs that anticipate fluences on the high side of this equation will likely be dominated by LID considerations. For fluences below this range, one can reasonably assume that LID will not be a major factor limiting system performance.

Note that equation 1 has little theoretical basis except for the case where damage is caused by a thin absorbing film on a surface of interface. For that case, damage fluence should scale as the square root of the pulse width due to one-dimensional heat dissipation into the surface.

If one must know the damage threshold to better than the approximation in Equation 1, then one should make a careful measurement of the threshold for conditions (tp, wavelength, etc.) similar to those expected in use.

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