

# Laser-induced damage and the role of self-focusing

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**Abstract.** We review the influence of self-focusing on the measurement of bulk laser-induced-damage (LID) thresholds in normally transparent optical materials. This role is experimentally determined by measuring the spot size and polarization dependence of LID and by observing beam distortion in the far field. Utilizing these techniques, we find that by using a tight focusing geometry in which the breakdown power is below  $P_2$ , the effects of self-focusing can be practically eliminated in an LID experiment.  $P_2$  is the so-called second critical power for self-focusing, and  $P_2 = 3.77P_1$ , where  $P_1 = c\lambda^2/32\pi^2n_2$ , where  $c$  is the speed of light in vacuum,  $\lambda$  is the laser wavelength and  $n_2$  is the nonlinear index of refraction. This is in accordance with numerical calculations by J. H. Marburger [in *Progress in Quantum Electronics*, J. H. Sanders and S. Stenholm, eds., Vol. 4, Part 1, pp. 35–110, Pergamon, Oxford (1975)]. With this knowledge we determine that damage is only partially explained by avalanche ionization and that the initiation of damage is strongly influenced by extrinsic processes. We therefore conclude that we are measuring extrinsic LID.

*Subject terms:* laser-induced material modification; laser-induced damage; laser-induced breakdown; avalanche ionization; dielectric breakdown; multiphoton absorption; self-focusing.

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## 1. INTRODUCTION

In this paper we discuss bulk (as opposed to surface) laser-induced-damage (LID) in transparent optical materials, i.e., materials whose linear absorption at the input laser wavelength is the order of  $10^{-3}$  or less.<sup>1-3</sup> In anticipation of our conclusion that, with few possible exceptions, LID is influenced by extrinsic processes, we refer to the damage as extrinsic laser-induced-damage (ELID) in what follows. However, it is important to realize that how self-focusing affects damage threshold data is independent of the damage mechanism (i.e., intrinsic or extrinsic). By intrinsic we mean that the threshold is not increased by reducing the defect or impurity density. We investigate the role of self-focusing in LID and how the experimental geometry can alter this role. Misunderstandings concerning self-focusing effects have led researchers to incorrect conclusions concerning the observed parametric dependences of ELID, which in turn affect conclusions of the importance of extrinsic effects in LID. A clear understanding of the role of self-focusing in LID allows us to design experimental geometries in which such effects can be practically eliminated.

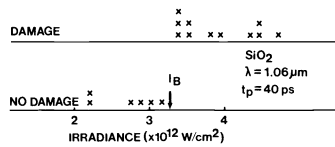


Fig. 1. Method of LID threshold determination. Damage (X on upper line) or no damage (X on lower line) for SiO<sub>2</sub>.

Parametric dependencies of ELID thresholds can then be unambiguously compared with predictions of theory. The overwhelming evidence from such studies points to the conclusion that damage in these transparent materials is not completely explained by avalanche ionization theory<sup>4,5</sup> but is heavily influenced by extrinsic effects. We give evidence supporting this conclusion by presenting ELID data for doped glasses and gamma-irradiated SiO<sub>2</sub>.

ELID is a threshold-like phenomenon. The threshold behavior is illustrated in Fig. 1, which shows a set of ELID data taken on a fused silica sample using 40 ps, linearly polarized, 1.06  $\mu\text{m}$  pulses focused to a Gaussian spot size of approximately 6.8  $\mu\text{m}$ . Throughout this paper pulsewidths are quoted as FWHM, and spot sizes are the HW  $1/e^2M$  in irradiance. The data are plotted as a function of internal irradiance with an X on the lower line if no damage occurred and on the upper line if damage did occur. The threshold is defined as the peak-on-axis fluence ( $\text{J}/\text{cm}^2$ ), irradiance ( $\text{W}/\text{cm}^2$ ), or rms electric field ( $\text{V}/\text{cm}$ ), which results in an irreversible change in the specimen as determined by one or more of the following: an increase in scattering of a HeNe probe beam (either by visual observation or photometric detection), a change in morphology as seen in a microscope, or a permanent change in the transmittance of the sample. In most cases this type of damage is obvious and all of the symptoms are observed. In addition, ELID in these materials is usually accompanied by a visual flash associated with the onset of dielectric breakdown (i.e., avalanche ionization). These data as well as other data discussed in this paper are one-on-one measurements in which each site is irradiated only once even if the site did not incur damage. It is observed in some materials that preirradiation with pulses below the damage threshold as defined can alter the ELID threshold, the so-called N-on-one effect. Note the very sharp threshold shown in Fig. 1, which is indicative of damage in transparent dielectrics. The threshold shown of approximately  $3.3 \times 10^{12} \text{ W}/\text{cm}^2$  is about two orders of magnitude larger than could be expected from the same sample used as an optical component in a system. This is the case since the failure mode for large illuminated areas used in practice is more likely surface damage where other extrinsic effects cause damage. In the experiments relevant to this paper, surface damage is avoided by focusing into the bulk of the material, keeping the irradiance at the damage-prone surfaces low.

The threshold-like behavior illustrated in Fig. 1 is indicative of an extremely nonlinear process. Considerable effort has been spent to monitor subthreshold changes in the optical properties of these materials with little success. A notable exception is the recent work of Bräunlich and coworkers at Washington State University.<sup>6-8</sup> They report the observation of multiphoton absorption of 532 nm light in carefully selected alkali halide samples of as high an order as four prior to damage using luminescence and the photoacoustic tech-

nique. We comment on the possible implications of their results in Sec. 5. No connection between the linear optical properties and the ELID thresholds has been found. Experiments have also shown that the transmittance of a laser pulse is cut off within a few picoseconds of the initiation of damage with no pulse distortion prior to damage.<sup>9</sup> This highly nonlinear behavior of ELID makes precise determination of the damage mechanism(s) difficult. In fact, we must infer the mechanism(s) of failure from parametric dependences (e.g., pulsewidth and wavelength dependence). This has resulted in slow progress in developing a complete understanding of the physical processes involved in ELID. On the other hand, as we shall see later in this paper, the first qualitative description of ELID was essentially correct.<sup>10,11</sup>

## 2. HISTORICAL REVIEW

The first reports of bulk ELID were by Bruma<sup>10</sup> and Hercher<sup>11</sup> in papers at the 1964 spring meeting of the Optical Society of America. The principal conclusions of these initial reports are as follows: (1) Linear absorption plays no major role in this type of failure. (2) The damage process is highly nonlinear. (3) Electron avalanche breakdown may be initiated by defects and/or impurities. (4) The ELID threshold and morphology depend on spot size. While there has been some controversy regarding the third statement, data now tends to confirm all of the statements made 25 years ago. Excellent sources for data on ELID in many materials are the proceedings of the annual Boulder Damage Conference starting in 1970 and the references therein.<sup>12</sup>

One can divide much of the past results in ELID experiments into two distinct categories. Category A consists largely of work conducted at Harvard University by Bloembergen<sup>1</sup> and his students Yablonovitch,<sup>13-15</sup> Fradin,<sup>16-20</sup> and Smith<sup>2,21-23</sup> and by their coworkers. The Category A experiments represent a systematic investigation of the breakdown thresholds of several alkali halides and fused quartz over wavelengths ranging from 10.6  $\mu\text{m}$  to 0.355  $\mu\text{m}$  and for pulse durations ranging from nanoseconds to tens of picoseconds. The results were interpreted in terms of an intrinsic model of electron avalanche breakdown.<sup>2</sup> The breakdown thresholds were found to vary little from sample to sample for a given material even if the samples were supplied by different crystal growers. The breakdown thresholds were found to agree within  $\pm 15\%$  over wavelengths ranging from 10.6  $\mu\text{m}$  to 1.06  $\mu\text{m}$ . It is important to note that this agreement in damage threshold for the alkali halides was obtained by comparing damage thresholds for different focusing conditions at the different wavelengths over a wide range of pulse durations and focusing conditions. In addition, the Category A workers found that the frequency dependence of the breakdown thresholds agreed with the theory of intrinsic avalanche ionization over the full wavelength range, although some evidence of multiphoton ionization was observed at 0.355  $\mu\text{m}$ .<sup>22</sup>

In part, the agreement of the results of the Category A workers with the predicted frequency dependence of intrinsic electron avalanche breakdown arose from attempts to correct the data at 1.064  $\mu\text{m}$ , 0.694  $\mu\text{m}$ , 0.532  $\mu\text{m}$ , and 0.355  $\mu\text{m}$  for the presumed effects of self-focusing. For example, Smith et al.<sup>21,22</sup> scaled the results of their picosecond breakdown work under the assumption that  $P_1$  (discussed in the next section) was the critical power of importance for focused Gaussian beams. They observed a slow increase in the scaled

thresholds with increasing frequency, consistent with the predictions of intrinsic avalanche theory. We now know  $P_1$  is not the critical power of importance in those experiments and the data should not have been scaled, thus invalidating the conclusions. The unscaled thresholds *decrease* with increasing frequency, opposite to the prediction of the intrinsic avalanche ionization theory.

On the other hand, the results of Category B workers were often found to be inconsistent with those of the Harvard group. Category B workers include Olness,<sup>24,25</sup> Yasojima and coworkers in Japan,<sup>26,27</sup> Bass and Barrett<sup>28</sup> and coworkers at the University of Southern California, Manenkov<sup>29</sup> and coworkers in the Soviet Union, Soileau,<sup>30,31</sup> Van Stryland,<sup>32</sup> and Sparks.<sup>4</sup> One of the main conclusions to be drawn from the Category B experiments is that laser-induced breakdown is caused by extrinsic properties of the material. In a study reported by Manenkov<sup>29</sup> in 1977, the breakdown irradiance at 1.06  $\mu\text{m}$  measured for 100 NaCl samples varied from sample to sample by a factor of 50. The highest thresholds reported in Manenkov's work were higher than previously reported "intrinsic" thresholds (e.g., three times those reported in Ref. 13). Similar results have been seen by Soileau<sup>30,31</sup> and in earlier studies by Olness<sup>25</sup> and Yasojima.<sup>27</sup> In addition, no systematic variation in the breakdown thresholds as a function of material bandgap was observed in the results of Olness<sup>24,25</sup> and Yasojima<sup>26,27</sup> in contrast to the Category A measurements. A decrease in the breakdown threshold with decreasing laser wavelength was seen by Soileau et al.<sup>30,31</sup> in the alkali halides over wavelengths ranging from 10.6  $\mu\text{m}$  to 1.06  $\mu\text{m}$ , as was observed for the unscaled thresholds of Smith et al.<sup>21,22</sup> Again, this is opposite to the trend predicted by intrinsic avalanche ionization theory. Finally, results in Category B experiments indicate the presence of a strong spot size dependence for the breakdown field in the alkali halides and fused quartz as well as other materials that could not be explained by self-focusing and is inconsistent with intrinsic avalanche breakdown theory. Also, the measured damage thresholds differed from sample to sample even in materials from the same manufacturer.

Much of the controversy found in the literature is due to the differences in the ways in which various authors account for, or attempt to account for, self-focusing. Early in the history of studies of ELID, self-focusing was recognized as one of the major contributors to catastrophic, irreversible changes in material properties.<sup>12</sup> Over the years many studies, both theoretical and experimental, have been conducted in attempts to account for self-focusing effects in bulk damage experiments. However, due to the complexity of the problem, agreement between theory and experiment has been mixed. While accurate, direct measurements of the nonlinear index of refraction,  $n_2$ , have been made using various techniques (e.g., interferometry<sup>33,34</sup>), the power for which significant changes occur in the linear propagation of focused Gaussian beams through nonlinear materials has only recently been established experimentally.<sup>35,36</sup> We present some of that work in what follows.

### 3. THEORY

#### 3.1. Self-focusing

Nonlinear refraction in a highly transparent dielectric results from a change in the index of refraction given by

$$\Delta n = n_2 \langle E^2 \rangle, \quad (1)$$

where  $\langle E^2 \rangle$  is the time-averaged square of the electric field and  $n_2$  is the nonlinear index of refraction. Here, we assume that the nonlinearity has a response time much less than the pulse duration. An alternative way of expressing the index change that has come into common use recently is  $\Delta n = \gamma I$ , where  $I$  is the irradiance and  $\gamma$  is a nonlinear index coefficient. These coefficients are related by a constant with<sup>33</sup>  $n_2$  (esu) =  $cn/40\pi \gamma$  (m<sup>2</sup>/W), where  $c$  is the speed of light in vacuum (m/s) and  $n$  is the linear index of refraction. Many mechanisms can give rise to self-focusing effects in solids. For tight focusing geometries using nanosecond and longer pulse durations, electrostriction, thermal self-focusing, and the electronic Kerr effect can all contribute to a catastrophic self-focus. For picosecond pulse durations the dominant mechanism in transparent solids is believed to be the electronic Kerr effect.

A large volume of work has been devoted to the study of self-focusing effects in solids.<sup>12</sup> The theories developed to describe the process indicate that self-focusing is dependent on the power of the laser beam in the material. Two critical powers of importance are often cited in the literature for Gaussian beams. The first of these,  $P_1$ , is given by<sup>37</sup>

$$P_1 = \frac{c\lambda^2}{32\pi^2 n_2}, \quad (2)$$

where  $\lambda$  is the laser wavelength. Many theories based on the constant shape approximation have assumed that a catastrophic collapse of the beam will occur in the material when the beam power approaches  $P_1$ . This idea was first introduced by Zverev and Pashkov,<sup>38</sup> who suggested the following equation for the irradiance enhancement due to self-focusing:

$$I_{\text{SF}} \approx \frac{I_0}{1 - P/P_1}, \quad (3)$$

where  $I_{\text{SF}}$  is the peak irradiance in the presence of self-focusing and  $I_0$  is the peak irradiance in the absence of self-focusing. This equation was subsequently used by researchers to "scale" damage data and "correct" for the effects of self-focusing.<sup>16,21,22</sup> Equation (3) can be rearranged to give the following equation:

$$\frac{1}{P} = \frac{1}{I_D} \left( \frac{1}{A} \right) + \frac{1}{P_1}, \quad (4)$$

where  $A$  is the focal beam area in the absence of self-focusing. Thus, a plot of inverse power for damage, versus inverse area, or spot size squared, was expected to yield both the critical power for self-focusing  $P_1$  as the intercept and the damage irradiance  $I_D$  from the slope. A critical assumption made in using this equation is that the damage irradiance is independent of focal area in the absence of self-focusing. We find this assumption invalid as discussed in Sec. 5.2.

In contrast to the earlier self-focusing studies, exact solutions of the nonlinear wave equation made by Marburger<sup>37</sup> have shown that even for focused geometries, significant deviations from normal linear propagation do not occur until the beam power exceeds  $P_1$  and a catastrophic collapse of the beam within the depth of focus does not occur until the beam

**TABLE I. Comparison of the constant shape approximation (CSA) to numerical solutions<sup>37</sup> (NS) for focused Gaussian beams.**

$P/P_2$	$l/l_0$ CSA	$l/l_0$ NS
0.27	1.37	1.30
0.60	2.50	2.08
0.80	5.00	3.94
0.90	10.00	7.15
0.95	20.00	16.45
0.96	25.00	28.60
0.97	33.30	63.40
0.98	50.00	100.00
0.99	100.00	192.00

power exceeds the second critical power  $P_2$ .  $P_2$  is defined as (for Gaussian beams)

$$P_2 = 3.77P_1 . \quad (5)$$

The factor of 3.77 in Eq. (5) comes from numerical solutions of the nonlinear wave equation for an input beam with a Gaussian spatial profile. The power  $P_2$  is the least power for a singular self-focus to occur within the Rayleigh range, i.e., the beam confocal parameter, for both prefocused and unfocused geometries. This means that for samples thicker than the Rayleigh range (as is the case for most bulk laser-induced damage experiments) a singular self-focus will occur *within* the sample, and LID will occur for an input power equal to  $P_2$ . Note that for tightly focused beams, i.e., very small spot sizes, the breakdown field will be reached before  $P$  approaches  $P_2$ . If that is the case, then LID results can be nearly independent of self-focusing effects. The small focal spot size needed will depend on the material,  $P_2$ , and the material breakdown threshold.

Marburger<sup>37</sup> found that the irradiance enhancement within the nonlinear material was given approximately by

$$I_{SF} \approx \frac{I_0}{1 - P/P_2} , \quad (6)$$

which is only valid for  $P \leq P_2/4$ . Note that this is identical to Eq. (3) with  $P_1$  replaced by  $P_2$ . When the input power is significantly greater than  $P_2/4$  one must use the more complete numerical solution to compute the enhanced irradiance due to self-focusing. Table I is a comparison of the approximation given by Eq. (6) and the exact numerical solution given by Marburger (see Ref. 37, p. 66). Note that while the enhancement cannot be calculated accurately by Eq. (6) for  $P > P_2/4$ , it still correctly predicts breakdown at a power of  $P_2$ .

In 1977 Smith et al.<sup>22</sup> also found some problem in using  $P_1$  for correcting their 532 nm and 355 nm picosecond damage thresholds for self-focusing. In most cases, their 355 nm thresholds were higher than  $P_1$ . A scaling factor was proposed such that the critical power varied between  $P_1$  and  $P_2$  depending on the value of  $P$  at damage. However, since the exact functional dependence for the intensification was unknown at the time, the thresholds reported in Ref. 22 included the breakdown power and the uncorrected focal area so that future workers could re-examine the data in the light of new measurements. We re-examine that data in Sec. 5.1.

We previously presented experimental evidence showing that, as predicted by Marburger, the important power for focused geometries is the second critical power,  $P_2$ .<sup>35,37</sup>

Indeed, Eq. (5) can be “verified” (i.e., we show that  $P_2$  is the critical power of importance) as shown in Sec. 4. Additional evidence for high  $P_2$  materials is given by measurements of the polarization dependence of the breakdown powers and measurements of beam distortions in the transmitted, time-integrated spatial beam profile discussed in Sec. 4.

### 3.2. Polarization dependence of self-focusing

The use of short pulses presents us with the advantage that we need only consider the fast electronic Kerr nonlinearity in data analysis. This nonlinearity is polarization dependent, and this polarization dependence presents us with a simple way of determining whether or not self-focusing effects are present in LID measurements.

Early papers in the literature have shown that the nonlinear refractive index for isotropic materials (such as fused quartz) is given by<sup>39–41</sup>

$$n_2 \text{ (L.P.)} = \frac{12\pi}{n} \chi_{1111}^{(3)} , \quad (7)$$

$$n_2 \text{ (C.P.)} = \frac{24\pi}{n} \chi_{1122}^{(3)} ,$$

for linearly polarized and circularly polarized light, respectively, where the  $\chi_{ijkl}^{(3)}$  are third-order nonlinear susceptibility tensor elements. A symmetry relation exists for isotropic materials such that

$$\chi_{1111}^{(3)} = 2\chi_{1122}^{(3)} + \chi_{1221}^{(3)} . \quad (8)$$

Measured values for these tensor elements indicate that for fused quartz  $\chi_{1122}^{(3)}$  is approximately equal to  $\chi_{1221}^{(3)}$ .<sup>41</sup> Thus, we can express  $n_2$  (C.P.) in terms of the same  $\chi^{(3)}$  tensor element as  $n_2$  (L.P.), giving

$$n_2 \text{ (C.P.)} = \frac{8\pi}{n_0} \chi_{1111}^{(3)} . \quad (9)$$

We see that the ratio of  $n_2$  for circular polarization to  $n_2$  for linear polarization is 2/3. This implies that the ratio of the critical powers for self-focusing for the two cases is 1.5.

A similar but slightly more complicated analysis for NaCl gives a ratio for the critical powers that varies between 1.37 and 1.46 depending on the propagation direction in the cubic crystal. We do not know the orientation of the large grain size crystalline samples and, therefore, expect a ratio between the above values near 1.4.

If we extend this concept to measurements of bulk optical breakdown and if self-focusing dominates the breakdown process, then, in both the isotropic and cubic cases, the ratios of the breakdown powers for the two polarization states should be equal to the ratio of the critical powers (i.e., 1.5 for SiO<sub>2</sub> and  $\approx 1.4$  for NaCl).

The polarization dependence of self-focusing has already been well established experimentally. For example, Moran et al.<sup>34</sup> measured  $n_2$  for various laser glasses (isotropic materials) using time-resolved interferometry and found that  $n_2$  (L.P.)  $\approx 1.5n_2$  (C.P.). Feldman et al.<sup>42</sup> measured the breakdown powers as a function of polarization for fused quartz and other glasses using nanosecond pulse durations at 1.06  $\mu\text{m}$ . He used the observed polarization dependence in an attempt to sepa-

**TABLE II.** The nonlinear refractive index  $n_2$  as measured using beam distortion<sup>43</sup> compared with other methods.

MATERIAL	WAVELENGTH $\mu\text{m}$	$n_2$ ( $\times 10^{13}$ esu)	$n_2$ (Others) ( $\times 10^{13}$ esu)
CS <sub>2</sub>	1.06	128 ± 30	125 ± 30 <sup>45</sup>
	0.53	123 ± 30	
NaCl	1.06	1.37 ± 0.30	1.22 ± 0.21 <sup>33</sup>
	0.53	1.38 ± 0.30	1.59 <sup>57</sup>
SiO <sub>2</sub>	1.06	0.62 ± 0.15	0.95 ± 0.10 <sup>33</sup>
	0.53	0.60 ± 0.15	0.85 <sup>47</sup>
BK-7	1.06	1.45 ± 0.30	1.46 ± 0.10 <sup>46</sup>
	0.53	1.01 ± 0.25	1.30 <sup>47</sup>

rate the various contributions to  $n_2$  and was the first to point out that the presence or absence of self-focusing in breakdown measurements could be determined by measuring the breakdown threshold power as a function of polarization. We use this concept in our own measurements to determine the contribution of self-focusing. The data presented in Sec. 4.2 on fused quartz and NaCl clearly show the transition from an experimental geometry in which self-focusing dominates the damage process to a geometry in which self-focusing can be neglected. In the next section we first verify Marburger's theory by monitoring breakdown in CS<sub>2</sub> at 1.06  $\mu\text{m}$  and 0.53  $\mu\text{m}$ .

## 4. EXPERIMENT

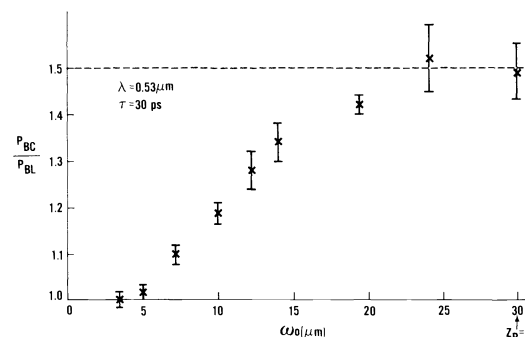
### 4.1. Experimental verification of Marburger's theory

Marburger's prediction<sup>37</sup> that breakdown will occur at an input power  $P_2$  given by Eq. (5) in a tight focusing geometry can be verified by arranging an experiment in which the breakdown threshold is very high and  $P_2$  is very low. A classic example of such a material is the liquid CS<sub>2</sub>. This material is an excellent choice for model system studies since its nonlinear behavior has been studied for years and is relatively well understood. The nonlinearity is due to nonresonant reorientation of the CS<sub>2</sub> molecules, which relaxes with a time constant of approximately 2 ps. The first step is to measure  $n_2$  in a manner independent of the laser-induced breakdown measurements. Table II is a summary of such measurements for CS<sub>2</sub> and other materials of interest using the beam distortion technique described in Ref. 43. Values obtained by other workers using various techniques are listed for comparison.<sup>33,44-47</sup> With the possible exception of SiO<sub>2</sub> (a 30% difference) the agreement with other methods is excellent. It is important to note that the observation of beam distortion in the far field as reported in Ref. 43 is performed at irradiance levels not far below damage, and the agreement with other data confirms the propagation analysis for high input powers.

We next set up a bulk breakdown experiment in CS<sub>2</sub>, i.e., arranged the sample length to be much longer than the confocal beam parameter, and measured the breakdown power.<sup>48</sup> We then used Eq. (5) to calculate  $n_2$  at both 1.06  $\mu\text{m}$  and 0.53  $\mu\text{m}$  and compared this with values obtained by beam distortion measurements<sup>44</sup> and time-integrated interferometry.<sup>45</sup> The results of this comparison are shown in Table III. In this experiment laser-induced breakdown is totally dominated by self-focusing and the breakdown power was experimentally determined to be independent of the focusing conditions. Note the excellent agreement between the  $n_2$  determined from the breakdown measurements using Eq. (5), which assumes

**TABLE III.** Comparison of the LID method using  $P_2$  with other methods of measuring  $n_2$  in CS<sub>2</sub>.

METHOD	$\lambda(\mu\text{m})$	$n_2 (\times 10^{11}$ esu)
Equation 5	1.06	1.3 ± 0.3 <sup>48</sup>
Equation 5	0.53	1.2 ± 0.3 <sup>48</sup>
Beam Distortion	1.06	1.5 ± 0.3 <sup>43</sup>
Beam Distortion	0.53	1.5 ± 0.3 <sup>43</sup>
Interferometry	1.32	1.3 ± 0.3 <sup>45</sup>



**Fig. 2.** A plot of the ratio of  $P_B$  for circularly polarized light to  $P_B$  for linearly polarized light as a function of spot size in SiO<sub>2</sub> (sample 79-FQ-7940-1). The dashed line represents the expected ratio when self-focusing dominates.

that  $P_2$  is the critical power, and those determined by beam distortion measurements. This verifies that the factor of 3.77 predicted by the theory in Ref. 37 is correct to within the error bars shown.

We have also experimentally determined when self-focusing is present in bulk breakdown experiments by observing the polarization dependence of damage as discussed in the following section.

### 4.2. Polarization dependence

Figure 2 is a plot of the ratio of the breakdown power for circular polarization to the breakdown power for linear polarization as a function of the focal spot radius measured in air.<sup>35,36</sup> The material is fused quartz, the laser wavelength is 0.53  $\mu\text{m}$ , and the pulse duration is 30 ps. Three regions of interest are clearly evident. For small focal radii and small breakdown powers the ratio  $P_{B\text{circular}}/P_{B\text{linear}}$  is approximately unity, indicating the lack of electronic self-focusing as discussed in Sec. 3.2. For focal radii greater than 23  $\mu\text{m}$  and large breakdown powers the ratio levels off to near the theoretical value of 1.5 indicating the dominance of self-focusing. The transition regime shows the data increasing from unity to the theoretical maximum, clearly showing the onset of electronic self-focusing in fused quartz.

Now, rather than looking at the ratio of the critical powers we examine the behavior of the breakdown powers directly in Fig. 3. Here, the breakdown power in megawatts is plotted as a function of the focal spot radius in air. The triangles represent the breakdown powers for linear polarization, and the circles are the breakdown powers for circular polarization. The horizontal dashed line represents the critical power  $P_2$  for linear polarization calculated from our measured  $n_2$  values for this sample.<sup>43</sup> As can be seen, the breakdown power for linear polarization increases with increasing focal radius and then reaches  $P_2$  for larger focal radii. In the region where the

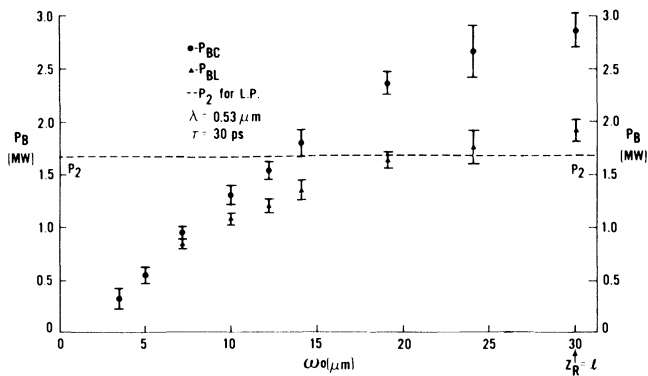


Fig. 3. A plot of the breakdown power versus spot size for the sample of Fig. 2 (triangles—linear polarization, circles—circular polarization). The dashed line gives  $P_2$  as calculated from the measured value of  $n_2$ .<sup>43</sup>

breakdown power becomes constant, the polarization ratio is approximately 1.5. Similar results are seen for fused quartz at  $1.06 \mu\text{m}$ . Both of these trends indicate that self-focusing dominates the breakdown process when the breakdown power approaches  $P_2$ .

Results similar to those seen for  $\text{SiO}_2$  are observed for NaCl. At  $0.53 \mu\text{m}$  for 30 ps pulses and small focal radii and therefore, small breakdown powers, the ratio is approximately unity. This indicates the lack of electronic self-focusing, whereas for large focal radii and large breakdown powers the ratio levels off near the mean theoretical value of approximately 1.4. In this region self-focusing dominates the breakdown process. Again, in the region where the breakdown powers remain constant with increasing spot size, the ratio of the breakdown powers reaches approximately 1.4. Similar results are also seen for this sample at  $1.06 \mu\text{m}$ . We discuss some of those measurements and their significance below.

At this point we remind the reader of the definition of the critical power for self-focusing and discuss more of Marburger's results.<sup>37</sup>  $P_2$  is the *least* power for which a catastrophic collapse will occur for both focused and unfocused geometries. The point of maximum on-axis irradiance does not occur at the beam waist; it occurs "downstream" of the beam waist at a distance comparable to the Rayleigh range in the material for  $P = P_2$ . If the sample thickness is thinner than the Rayleigh range, insufficient nonlinear material exists for self-focusing to cause a catastrophic collapse of the beam to occur in the material at  $P_2$ . For damage dominated by self-focusing, this means that the material will not fail until the beam power exceeds  $P_2$ . This type of behavior can be seen in the data presented in Fig. 4. Here we have plotted the breakdown power in megawatts as a function of the focal spot radius in air for the NaCl sample. The laser wavelength is  $1.06 \mu\text{m}$ , and the pulsewidth is 42 ps. The horizontal dashed line represents the critical power  $P_2$  calculated from measured  $n_2$  values of this sample (see Table II).<sup>43</sup> The vertical dotted line divides the data for which the sample thickness is less than the Rayleigh range (region to the right) from that in which the sample is thicker than the Rayleigh range (region to the left). Let us first examine only the triangular data points that are for a NaCl sample thickness of 2 in. If we examine the data for the region to the left, we see that the breakdown power increases with increasing focal radius to approximately  $P_2$ . However, when the sample becomes shorter than the depth of focus (region to the right), the breakdown power continues to

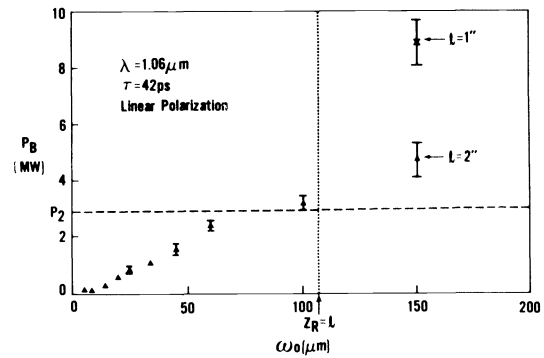


Fig. 4. Breakdown power as a function of spot size in NaCl for a 2 in. thick sample (triangles) and a 1 in. thick sample (X). The horizontal dashed line gives  $P_2$  as calculated from  $n_2$ ,<sup>43</sup> and the vertical dotted line gives the spot size at which the Rayleigh range of the focused beam equals the sample thickness for the 2 in. sample.

increase. If we now rotate the sample so that the beam propagates through 1 in. of material instead of 2 in., a dramatic increase in the breakdown power is observed. This is seen by examining the data points for the  $150 \mu\text{m}$  spot size. The triangular point is for a sample thickness of 2 in., and the point represented by the X is for 1 in. of material. No other parameters have been changed. To show that the change in the breakdown power was not due to an orientation effect a similar test was performed for a case in which the sample was thick compared with the Rayleigh range for 1 in. of material. No change in the breakdown power was observed when the sample was rotated. This simple test clearly shows the effects of self-focusing in breakdown measurements.

## 5. CONCLUSIONS FROM THE ROLE OF SELF-FOCUSING IN LID

### 5.1. Re-evaluation of ELID data

The significance of this work should be emphasized. Much of the early experimental work on self-focusing used the scaling law proposed by Zverev et al.<sup>38</sup> to correct the data for self-focusing. In that work the critical power of importance was assumed to be  $P_1$ . Spot size dependencies of the laser-induced damage thresholds were assumed to be merely a reflection of the effect of self-focusing since the breakdown powers were in most cases a fair fraction of  $P_1$ . However, since the self-focusing theory predicts and this work confirms that the critical power is in fact  $P_2$  and not  $P_1$ , the spot size dependencies are in most cases due to other mechanisms in the material, possibly the contribution of defects to the LID thresholds. Work that used the method of Zverev et al. needs to be re-examined.

Unfortunately, much of the work using the Zverev and Pashkov<sup>38</sup> scaling is not recoverable from the literature since the uncorrected thresholds are not reported and cannot be extracted due to insufficient information. However, Fradin et al.<sup>19</sup> in anticipation of some problem with the Zverev and Pashkov method did not scale the data reported in 1973 for self-focusing, and in retrospect they were correct in not scaling the data. They observed that the damaging power in NaCl scaled as the square of the focal length of the lenses used for the picosecond pulsewidths. As noted earlier, in 1977 Smith et al.<sup>22</sup> also found some problem in using  $P_1$  to correct their 532 nm and 355 nm picosecond damage thresholds for self-focusing. They included the breakdown power and the uncorrected focal area in their report so that future workers could re-

**TABLE IV. Breakdown thresholds at 0.53  $\mu\text{m}$  of Ref. 22 scaled for self-focusing using  $P_1$  (column 7) and  $P_2$  (column 8).  $P$  stands for the breakdown power  $P_B$ ,  $E_0$  is the breakdown field, and  $A$  is the focal area calculated using linear optics.**

Material	$P_B$ (KW)	$A$ ( $\mu\text{m}^2$ ) (unscaled)	$P/P_1$	$P/P_2$	$E_0$ (MV/cm) (unscaled)	$E_{P_1}$ (scaled)	$E_{P_2}$ (scaled)
$\text{KH}_2\text{PO}_4$	151.0	16.0	0.57	0.15	15.3	23.4 (53%)	16.6 (8%)
$\text{SiO}_2$	129.0	15.9	0.46	0.12	14.5	19.0 (31%)	15.4 (6%)
$\text{NaCl}$	38.4	15.1	0.60	0.05	7.9	12.4 (57%)	8.1 (3%)
$\text{CaF}_2$	146.0	15.9	0.62	0.09	15.5	25.2 (63%)	16.3 (5%)
$\text{NaF}$	126.0	15.8	0.45	0.05	15.0	19.4 (30%)	15.4 (3%)
$\text{LiF}$	171.0	16.1	0.59	0.06	16.9	26.5 (57%)	17.5 (4%)

examine the data in the light of new measurements.

We can now use Eq. (5) to re-examine the 532 nm breakdown data of Smith et al.<sup>22</sup> In that work, the breakdown powers are all below  $P_1$ , so the irradiance increase predicted by Eq. (5) should be valid. Six materials were studied, including fused quartz and NaCl, as shown in Table IV. In examining the data we find that the breakdown threshold fields increased as much as 50% to 60% using  $P_1$ . These increased thresholds were reported. However, when the thresholds are properly corrected using the second critical power  $P_2$  we find on the average only a 5% increase. This is well within the  $\pm 15\%$  absolute uncertainty in the measurements.

Smith et al.<sup>22</sup> also reported breakdown thresholds at 355 nm for three of the materials listed in Table IV. The results for this near-UV study indicate that, in most cases, the breakdown threshold powers for these materials were substantially higher than the  $P_1$  critical powers at this wavelength. The only exception was  $\text{CaF}_2$  where  $P_B$  was found to be  $0.7P_1$ . In an attempt to correct their data for the presence of self-focusing they scaled their breakdown threshold irradiance levels in  $\text{KH}_2\text{PO}_4$ ,  $\text{LiF}$ , and  $\text{SiO}_2$  using Eq. (5) but with a critical power somewhere between  $P_1$  and  $P_2$ .

While they were on the right track, meaningful comparison of the 355 nm data with results at other wavelengths is difficult due to the reported poor spatial quality of the 355 nm beam used in the measurements. The uncertainty in the energy distribution within the focal area lead Smith et al.<sup>22</sup> to assign a factor of two range for the scaled breakdown threshold fields at this wavelength. The actual breakdown thresholds may or may not be within this range. A further complicating factor for interpreting the 355 nm work is the recent result that the effective  $n_2$  for a material may not be constant as a function of wavelength for photon energies approaching a substantial fraction of the band-gap energy. Based on our measurements using the technique of Ref. 43 in BK-7 (see Table II comparing  $n_2$  at 1  $\mu\text{m}$  and 0.5  $\mu\text{m}$ ) and those of White et al.<sup>49</sup> in BK-10 at 355 nm, the  $n_2$  values for the three materials studied at the third harmonic wavelength may well be substantially lower than the values at 1064 nm. This points to the need for accurate measurements of  $n_2$  in these materials in the regime where multiphoton effects may be coming into play. It may also be necessary to measure nonlinear refraction at near-damaging irradiance levels as is done in the beam distortion method.<sup>43</sup> This being the case, we will not attempt at this time to reexamine the 355 nm thresholds.

In our re-evaluation of the breakdown results of other workers, we have concentrated on the work of Ref. 22 for two reasons. The first reason is that the breakdown measurements in that work were conducted for pulse durations comparable to our own. Therefore, the self-focusing mechanisms in the test materials will be the same. The second reason is that, of the experimenters who scaled their breakdown thresholds for the presumed presence of self-focusing, Smith et al.<sup>22</sup> compose one of few groups whose work contains sufficient information and experimental parameters to determine the true breakdown thresholds. Other workers merely reported the scaled breakdown threshold irradiance levels without including the focal spot radii used in the measurements. This makes it impossible to recalculate the breakdown thresholds. In addition, the incorrectly scaled data of Smith et al.<sup>22</sup> was used in support of the intrinsic avalanche ionization model.

Several other breakdown studies have been conducted in these materials in which no self-focusing corrections were made. For example, Manenkov<sup>29</sup> reported breakdown measurements in the alkali halides (including NaCl) for nanosecond pulse durations at 10.6, 1.06, 0.69, and 0.53  $\mu\text{m}$  laser wavelengths. There is some uncertainty in the focal spot radius used in the measurements since two values are reported without specifying which correspond to the breakdown irradiance levels listed in Manenkov's work.<sup>29</sup> Feldman et al.<sup>42</sup> estimate that electrostriction is of relatively minor importance for the nanosecond pulses used in these experiments. However, we will use the larger value of  $n_2 = 4 \times 10^{-13}$  esu calculated from nanosecond three-wave mixing experiments.<sup>50</sup> With this in mind we find that  $P_B$  in NaCl is  $0.5P_2$  at 532 nm and  $P_B$  is  $0.14P_2$  at 1064 nm if we use the larger of the two focal radii cited in Ref. 29. If we use the small focal radius we find that  $P_B$  for NaCl is  $0.07P_2$  at 532 nm and  $0.02P_2$  for 1064 nm. Thus, self-focusing effects in Manenkov's<sup>29</sup> work for NaCl are negligible except perhaps for the combination of the largest spot size and shortest wavelength and, therefore, should not be scaled. The wavelength dependence reported by Manenkov from 10.6  $\mu\text{m}$  to 0.69  $\mu\text{m}$  was an increase in threshold much stronger than that reported by Fradin<sup>17</sup> (a factor of three). However, the threshold dropped at 0.53  $\mu\text{m}$  for a net drop going from 1  $\mu\text{m}$  to 0.5  $\mu\text{m}$  consistent with our measurements.

In a similar nanosecond study, Merkle et al.<sup>51-53</sup> reported single shot damage thresholds for Corning 7940 fused quartz for laser wavelengths ranging from 1064 nm to 355 nm. In fused quartz as well as in NaCl, for the tight focusing geometry used, electrostriction has been shown to play a small role in self-focusing effects for pulse durations around 30 ns.<sup>42</sup> We use the  $n_2$  value of  $0.95 \times 10^{-13}$  esu reported by Feldman et al.<sup>42</sup> for nanosecond pulse durations in  $\text{SiO}_2$ . We find that the breakdown powers reported by Merkle et al.<sup>51</sup> for Corning 7940 were less than  $0.08P_2$  at 1064 nm and equal to  $0.07P_2$  at 532 nm. Therefore, self-focusing effects in the work of Merkle et al.<sup>51</sup> are negligible. They report a decrease by a factor of two in threshold in going from 1.06  $\mu\text{m}$  to 0.53  $\mu\text{m}$ .<sup>51</sup>

We have also re-examined our own results published in Refs. 3 and 32. In Ref. 3 we used beam distortion and polarization dependence to verify that self-focusing was not the dominant breakdown effect. However, these tests (i.e., polarization dependence and beam distortion) were not conducted for each experimental condition used. For the most part, little (a few percent) or no adjustment of the originally published numbers is needed. For the ultra-short pulse data (pulsewidth



less than 5 ps) and largest spot size of 14  $\mu\text{m}$  for  $\text{SiO}_2$  at 1054 nm, adjustments as high as a factor of two were needed. These corrections do not change any of the trends observed or conclusions drawn from that data as discussed in the next section.<sup>3,32</sup> In addition, we should note that we have studied many samples of these materials grown by various techniques over the past 15 years. While the thresholds vary from sample to sample, from boule to boule, and even from different samples taken from the same boule, the data presented here is consistent with data obtained from similar “good” optical quality material. We have not found an exceptional sample that behaved differently from the behavior of the samples reported here.

## 5.2. Wavelength, temporal, and spot size dependence

Given that we now can account for the effects of self-focusing in an LID experiment, we can unambiguously determine the dependence of the threshold on various parameters such as pulsewidth, spot size, and wavelength. The conclusion reported by Smith et al.<sup>22</sup> that the observed wavelength dependence agreed with the predictions of avalanche ionization theory depended on the scaling using Eq. (3) with  $P_1$  as the critical power. Using  $P_2$ , the data of Ref. 22 shows a decrease in threshold with decreasing wavelength, which is not consistent with intrinsic avalanche breakdown. We find similar results for the wavelength dependence. While this wavelength dependence is in the direction predicted by intrinsic multiphoton-induced damage,<sup>54</sup> the dependence is much too weak.

As the order  $m$  of the multiphoton absorption process is increased, the irradiance needed to obtain the same absorptance increases approximately as the inverse ratio of the nonlinear absorption coefficients.<sup>55</sup> This ratio is estimated by Wherrett<sup>55</sup> to be  $\approx 10^{-4}I$ , where  $I$  is the irradiance. Experiments indicate that this ratio is  $\approx 10^{-3}I$  (Refs. 56 and 58). For example, the two-photon absorption coefficient of CdS at 0.53  $\mu\text{m}$  is reported<sup>56</sup> as 5.5  $\text{cm}/\text{GW}$ , and its three-photon absorption coefficient at 1.06  $\mu\text{m}$  is reported<sup>58</sup> as 0.01  $\text{cm}^3/\text{GW}^2$ . Thus, to obtain the same absorptance at the two different wavelengths would require an irradiance of  $\approx 550$   $\text{GW}/\text{cm}^2$  (or more, theoretically). Four-photon absorption would require a correspondingly higher irradiance. From 10  $\mu\text{m}$  to 0.5  $\mu\text{m}$  the experimentally measured irradiance thresholds change but only by factors of two to four. The reported observation of four-photon absorption at 532 nm prior to damage by the group at Washington State University is interesting and may indicate that intrinsic multiphoton absorption is responsible or partially responsible for damage in the selected samples of NaCl studied by that group. We note, however, that the high irradiance used by the WSU group of 550  $\text{GW}/\text{cm}^2$  for picosecond pulses is  $\approx 5.5$  times the damage threshold observed for the “best” samples reported by Manenkov using nanosecond pulses.<sup>29</sup> The absorptance due to direct four-photon absorption would then be of the order of  $5.5^{-3}$ , which is  $\approx 6 \times 10^{-3}$ , lower for the nanosecond pulses. It seems unlikely that if damage for picosecond pulses is caused by intrinsic multiphoton absorption that the same mechanism is responsible for the nanosecond data of Manenkov.<sup>29</sup> Gorshkov et al.<sup>59</sup> also found inconsistencies with the avalanche model in the temperature dependence of damage of the alkali halides at 0.53  $\mu\text{m}$ . We agree with the authors of Ref. 8 that “insufficient and contradictory data exist for  $m = 4$  in NaCl ( $E_g = 8.6$  eV,  $h\nu = 2.33$  eV) to unequivocally assign

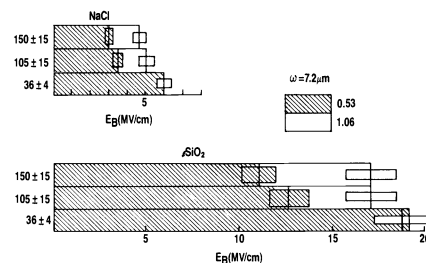


Fig. 5. Wavelength dependence of  $E_B$  for NaCl and  $\text{SiO}_2$  for several pulsewidths at 1.06 and 0.53  $\mu\text{m}$ . These thresholds have been scaled by a few percent for self-focusing using  $P_2$  and measured values of  $n_2$ .<sup>43</sup>

damage to the avalanche mechanism.” However, since the damage threshold varies considerably from sample to sample, we must still conclude that extrinsic effects are present and dominant for the lower threshold samples. Additionally, intrinsic multiphoton absorption may contribute carriers for an eventual avalanche, although no evidence for this was observed in the photoacoustic data of Ref. 8. At 0.5  $\mu\text{m}$  in these wide-gap materials the question of the role of intrinsic multiphoton absorption in LID remains. However, it is unlikely that for the wavelengths of 1  $\mu\text{m}$  and longer intrinsic multiphoton absorption is playing a role.

We reported damage on 13 samples of NaCl at 1  $\mu\text{m}$  and 0.5  $\mu\text{m}$  in Ref. 3. While the field thresholds varied from sample to sample by as much as a factor of four, we saw no systematic differences in the parametric dependencies of the ELID thresholds, although not all experiments were performed for all samples. Figure 5 shows a bar graph of damage in NaCl (upper) and  $\text{SiO}_2$  (lower) for a focal spot size in air of 7.2  $\mu\text{m}$  for various pulsewidths as indicated, for both 1.06  $\mu\text{m}$  (unshaded) and 0.53  $\mu\text{m}$  (shaded). The small corrections for self-focusing have been included in this figure. Except for the shortest pulses in both NaCl and  $\text{SiO}_2$  samples, the ELID threshold is reduced in going from a wavelength of 1  $\mu\text{m}$  to 0.5  $\mu\text{m}$ , which is inconsistent with an intrinsic avalanche model.

In the avalanche breakdown model of Refs. 4 and 5 the ionization rate is proportional to  $E^2$  in the high field limit (i.e., short pulse limit). Then, the buildup of carriers is given by

$$N = N_0 \exp(aE^2 t), \quad (10)$$

where  $N$  is the carrier density,  $N_0$  is the initial carrier density, and  $a$  is a material dependent constant. This limit corresponds to the situation in which the increase in energy of the electrons in the conduction band is simply proportional to the input irradiance and that all losses are negligible. This says that the ionization rate is limited by the rate at which the input light beam can supply energy to the conduction band electrons. It is commonly assumed that damage occurs when the carrier density reaches a critical value  $N_c$ . Thus, Eq. (10) can be solved for the breakdown field showing a  $t_p^{-0.5}$  dependence:

$$E_B = \frac{1}{\sqrt{at_p}} \ln^{1/2} \left( \frac{N_c}{N_0} \right). \quad (11)$$

We find that for very short pulses for NaCl this pulsewidth dependence is valid and the breakdown fluence is constant, while for  $\text{SiO}_2$  we see a somewhat weaker  $t_p^{-0.3}$  dependence.



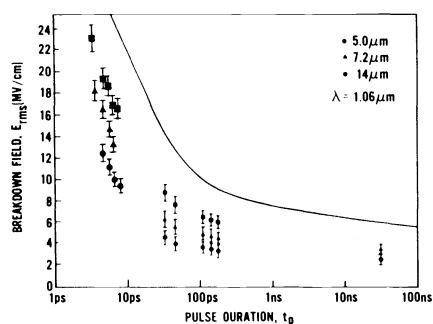


Fig. 6. Pulsewidth dependence of the rms breakdown field  $E_B$  for NaCl at  $1.06 \mu\text{m}$  for three different spot sizes. Data are scaled for self-focusing using  $P_2$ .<sup>43</sup> The solid line was obtained from the electron avalanche ionization theory of Sparks et al.<sup>4</sup>

For the low field limit, i.e., longer pulses, the ionization rate is exponentially dependent on  $E$  and the resulting pulsewidth dependence is relatively weak.<sup>4</sup> In Fig. 6, we reproduced the theoretical curves derived by Sparks et al.<sup>4</sup> showing the predicted dependence of the breakdown field  $E_B$  on pulsewidth and have extended the pulsewidth scale to longer and shorter pulses. The overall dependence of the data for a given spot size is in remarkably good agreement with this theory using no adjustable parameters. However, intrinsic avalanche ionization does not predict a spot size dependence, which is clearly seen in the data. This relatively large spot size dependence may well be due to extrinsic materials parameters such as multiphoton ionization of impurities or defects that provide the starter electrons for the avalanche  $N_0$ .<sup>3,32</sup>

While the observed temporal dependence is consistent with an intrinsic avalanche, the wavelength and spot size dependence strongly indicate that the phenomena is an *extrinsic* property of materials. In addition, the strong sample-to-sample variations in thresholds support this conclusion.<sup>12,29</sup> The extrinsic nature of bulk damage makes quantitative, first principle descriptions of the damage process very difficult. One problem is that the good optical materials of interest have very low levels of defects and impurities. One solution to this problem is to do experiments with samples prepared with known type and densities of defects and impurities. This is the approach we take below.

## 6. CONTROLLED DEFECT STUDIES

The effects of  $\gamma$  irradiation (which produces, among other defects,  $E'$  centers)<sup>60</sup> on the ELID thresholds of fused silica, were investigated. Samples were sliced into four quadrants. One quadrant was kept as a control (i.e., no irradiation), and the remaining three quadrants were subjected to  $10^6$ ,  $10^7$ , and  $10^8$  rad of cobalt  $\gamma$  irradiation from the Naval Research Laboratory cobalt source. Unfortunately, the density of defects cannot be determined,<sup>60</sup> although their effects on linear absorption and ELID can be measured. Single specimens of three types of fused silica were tested: Spectrasil A, B, and WF (water free). Damage was performed with 18 ns (FWHM) pulses at  $1.06 \mu\text{m}$  and  $0.53 \mu\text{m}$  focused into the bulk of three 3 mm thick samples using a 40 mm focal length lens. Each site was irradiated only once. The ELID thresholds were found to be independent of  $\gamma$  irradiation at the  $1.06 \mu\text{m}$  wavelength. However, significant reduction in damage thresholds (up to 40%) was measured for the irradiated samples at  $0.53 \mu\text{m}$ .<sup>61,62</sup> Again, the effects of self-focusing were determined to be negligible by performing polarization dependent damage mea-

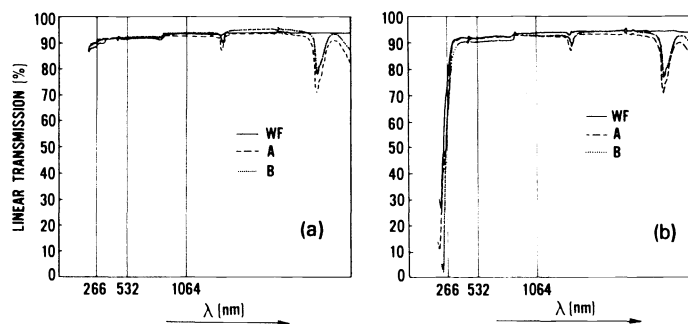


Fig. 7. Transmission as a function of wavelength  $\lambda$  for the three samples of SiO<sub>2</sub> (a) unirradiated and (b)  $\gamma$  irradiated with  $10^8$  rad.

TABLE V. Composition of the various fluorozirconate glasses studied.

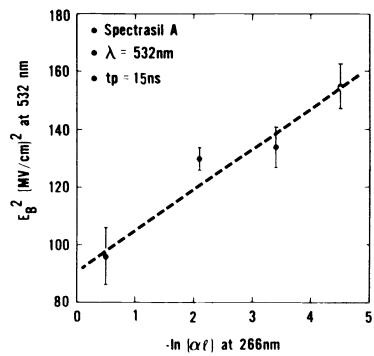
Sample	Color	ZrF <sub>4</sub> (mol%)	BaF <sub>2</sub> (mol%)	LaF <sub>3</sub> (mol%)	AlF <sub>3</sub> (mol%)	CrF <sub>3</sub> (mol%)	MnF <sub>2</sub> (mol%)	NiF <sub>2</sub> (mol%)	NdF <sub>3</sub> (mol%)
A	Clear	57	34.5	4.5	4				
B	Green	56.75	34.25	4.5	4	0.5			
C	Green	56.5	34	4.5	4	0.5	0.5		
D	Green	56.5	34	4.5	4	0.5			0.5
E	Violet	55.75	33.75	4.5	4		1		1
F	Yellow	56.75	34.25	4.5	4			0.5	

surements. The ELID thresholds at  $0.53 \mu\text{m}$  are substantially lower than those at  $1.06 \mu\text{m}$  even for the unirradiated witness samples. Avalanche ionization predicts an increase in threshold for reduced wavelength. Thus, the ELID process is not simply avalanche breakdown even for the witness sample.

Figure 7 shows the linear transmission spectra of the unirradiated and irradiated ( $10^8$  rad) samples.<sup>61,62</sup> Note that this figure shows no observable change in transmission at either  $1.06 \mu\text{m}$  or  $0.53 \mu\text{m}$  but does show a substantial change at  $0.266 \mu\text{m}$  for the irradiated samples. This suggests that the decrease in ELID threshold at  $0.53 \mu\text{m}$  may be associated with the change in transmission at the harmonic  $0.266 \mu\text{m}$ . We will examine this relation after presenting data that shows a similar dependence of ELID on linear absorption at a harmonic.

We also performed a series of LID experiments on fluorozirconate (FZ) glasses, undoped and doped with Cr, Mn, Ni, and Nd.<sup>63</sup> Table V shows the glass compositions. The linear absorption spectra are given in Ref. 63. The doped and undoped FZ glasses were studied at  $1.06 \mu\text{m}$  using 45 ps and 18 ns pulses and a calculated  $5.3 \mu\text{m}$  focused spot size. Measurements at  $0.53 \mu\text{m}$  were performed using 18 ns pulses and a calculated focused spot size, assuming negligible aberrations, equal to  $2.7 \mu\text{m}$ .<sup>63</sup> Polarization dependent studies confirmed the absence of self-focusing effects. The breakdown fields decreased significantly at both wavelengths for all of the doped FZ glasses when compared with the undoped sample A.

We present below a two-photon assisted electron avalanche breakdown model as given in Refs. 62 and 63 that we justify primarily on the basis of its surprisingly good fit of the data. Our experimental results for both the irradiated silica and FZ glass materials were found to be in good agreement with this very simplified model in which two-photon absorption by impurities or defects provides the starter electrons for an avalanche. In this model we assume that laser-induced breakdown occurs when the density of free carriers generated by a combination of two-photon and avalanche processes



**Fig. 8.** Square of the damaging electric field at 532 nm versus the negative natural logarithm of the linear absorption at 266 nm for the irradiated and unirradiated Spectrasil A SiO<sub>2</sub> samples.

reaches some critical value  $N_c$ . In the avalanche process the buildup of free carrier density is given by Eq. (10), where we assumed the high field limit with an ionization rate proportional to  $E_B^2$ . The model assumes that the initial free carriers are generated by two-photon absorption from impurity states within the bandgap. Therefore,  $N_0$  can be written as  $N_0 \propto n$ , where  $n$  is the density of two-photon allowed impurity or defect states. Taking the natural logarithm of Eq. (10) yields

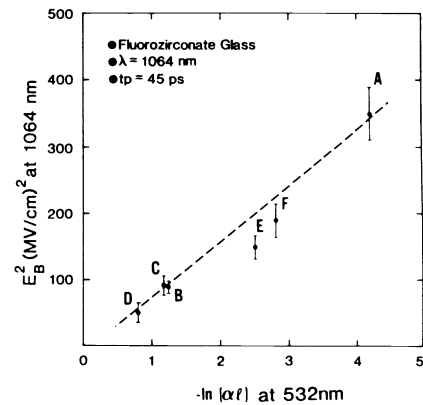
$$aE_B^2 = -\ln(n) + \text{constant} \quad (12)$$

The final assumption is that the density of two-photon allowed states at the fundamental wavelength ( $\lambda$ ) is proportional to the linear absorption at the second harmonic ( $\lambda/2$ ). For that assumption Eq. (12) gives

$$E_B^2|_{\text{at } \lambda} \propto -\ln(\alpha\ell)|_{\text{at } \lambda/2}, \quad (13)$$

where  $\alpha$  is the linear absorption coefficient at the second harmonic wavelength and  $\ell$  is the sample length.

Figure 8 is a plot of the square of the breakdown field at 532 nm as a function of  $-\ln(\alpha\ell)$  at 266 nm for the Spectrasil A sample. Note that increasing irradiation dosage goes from right to left in this figure. A linear relation is also seen for the other samples (B and WF), although sample B shows a significant increase in absorption for only the highest  $\gamma$  irradiation level. Figure 9 is a plot of the square of the breakdown field at 1.06  $\mu\text{m}$  versus  $-\ln(\alpha\ell)$  at 0.53  $\mu\text{m}$  using picosecond pulses for the undoped and doped FZ glasses. A similar plot for nanosecond pulses also yields a linear relation. Plotting the breakdown field squared at 0.532  $\mu\text{m}$  versus the  $-\ln(\alpha\ell)$  at 0.266  $\mu\text{m}$  using nanosecond pulses again gives a linear relation. Similar plots of damage versus linear losses at the damaging wavelength show *no* systematic trends. The agreement with the prediction of Eq. (13) is quite good considering the extreme simplicity of the model and the assumptions made. Note that even in cases in which nanosecond pulses were used where the ionization rate is expected to depart from the  $E_B^2$  dependence good fits are obtained. At what pulsewidth or irradiance the deviation from an  $E_B^2$  dependence becomes large is not well established. However, we found that for all of the FZ glasses where both nanosecond and picosecond data were taken, the reduction in ELID threshold for the doped samples was nearly a factor of two larger for the picosecond irradiation. This trend is expected in this model where the starter electrons for an avalanche are created by nonlinear



**Fig. 9.** Square of the damaging electric field at 1064 nm versus the negative natural logarithm of the linear absorption at 532 nm for the Fluorozirconate glass samples listed in Table V.

absorption since the breakdown field is substantially larger for picosecond pulses.

In spite of the complexity associated with laser-induced damage mechanisms for dielectric materials, our experimental results are consistent with a two-photon-assisted electron avalanche process. We should note that if this model is correct, the implication is that the witness samples at 0.53  $\mu\text{m}$ , in the case of the fused silica samples, and the undoped glass (sample A) in the case of the fluorozirconate glass samples are also dominated by extrinsic defect (or impurity) initiated ELID. This conclusion comes from the fact that these samples are included in the curves of ELID versus  $-\ln$  absorption, and they fit. Just how far we can take this simple model is not clear. The primary justification in presenting it is that the data fits the prediction. One conclusion, however, is certainly true; the damage in these materials is dependent on defects and impurities.

## 7. CONCLUSION

We have clarified the role of self-focusing in laser-induced-damage experiments and can readily account for its effects on measured thresholds. With this knowledge we have carefully examined both our data and the data of others to determine that intrinsic avalanche ionization theory cannot account for the parametric dependencies observed. Sample-to-sample variations, wavelength dependence, and spot size dependence of damage thresholds strongly suggest that damage to transparent dielectric materials is an extrinsic process. In a sense, this is good news in that the implication is that ELID thresholds can be increased as progress is made in the materials growth and preparation areas.

## 8. ACKNOWLEDGMENTS

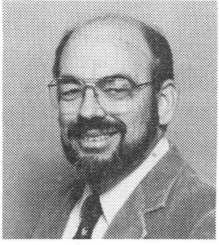
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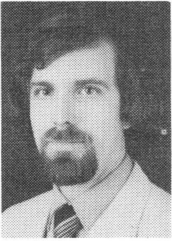
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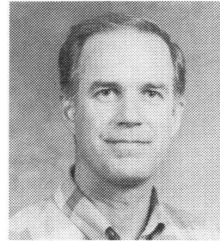
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