

## Self-Focusing in Damage Experiments Revisited

M. J. Soileau, William E. Williams,\* and Eric W. Van Stryland

Center for Applied Quantum Electronics  
Department of Physics  
North Texas State University  
Denton, Texas 76203

Last year we reported that the power of importance in self-focusing experiments is the second critical power,  $P_2$ , defined by  $P_2 = 3.77 P_1$  rather than the often quoted  $P_1 = c\lambda^2 / (32\pi^2 n_2)$ . Here  $n_2$  is the nonlinear refractive index,  $c$  the speed of light and  $\lambda$  the wavelength (both in vacuum). The factor of 3.77 is a numerical factor coming from computer calculations as discussed by Marburger [1]. We also presented a method by which we could obtain  $n_2$  at irradiances very near to damage. In addition, we showed the experimental conditions under which self-focusing is unimportant in damage experiments. We have now extended these results so that in damage experiments where self-focusing is important we can estimate the reduced spot size within the bulk at damage and, thus, obtain the damaging electric field magnitude. This we do by observing the far field time integrated spatial irradiance distribution. We present data for  $\text{SiO}_2$ ,  $\text{NaCl}$ , BK-7, and  $\text{CS}_2$ .

Key Words: critical power; laser damage; picosecond pulses; self-focusing.

### 1. Introduction

The role of self-focusing in laser-induced breakdown was examined in detail in a paper presented at the 1983 Boulder Damage Symposium [2]. In this paper we address questions raised in response to that paper and present additional data that support the conclusions reached in reference 2. The conclusion of reference 2 is that the critical power of importance for laser-induced damage experiments employing tightly focused Gaussian beams is the second critical power  $P_2$ , where

$$P_2 = 3.77 P_1 \quad (1)$$

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

where  $\lambda$  is the wavelength,  $c$  is the speed of light, and  $n_2$  is the nonlinear refractive index. (Equations (1) and (2) are taken from the classic work of Marburger [1].) We based this conclusion upon measurements of the polarization dependence of picosecond laser-induced breakdown and upon measurements of beam distortions in the time-integrated spatial profile of beams transmitted through the samples. In a related paper we reported the results of  $n_2$  in  $\text{NaCl}$  and  $\text{SiO}_2$  at 1064 nm and 532 nm [3].

Much of the past bulk laser-induced damage research assumed that self-focusing corrections to damage data are required for data taken with input power a fraction of  $P_1$  [4]. On the other hand, we have published work for which no self-focusing corrections were made even though the measured breakdown power was a substantial fraction of  $P_1$  [5,6]. In references 5 and 6 we used the lack of polarization dependence and lack of observable beam distortions to justify neglecting self-focusing in computing the laser-induced breakdown electric fields. It is critical that researchers interested in this problem take into account these differences in interpretation and data reduction when comparing the results and conclusions of the various bulk laser-induced

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\*Present Address: Texas Instruments, Inc., 8505 Forest Lane, P.O. Box 660246, M/S 3123, Dallas, Texas 75266

breakdown experiments. One can question the sensitivity of the null test used in references 5 and 6 in ruling out self-focusing. For example, the beam distortion measurements involve the time integrated spatial profile and are only sensitive to about  $\lambda/4$  peak distortion in the transmitted beam. This problem emphasizes the importance of direct measurements of  $n_2$  in a manner independent of the damage test.

In the sections that follow we will review the theoretical treatment of self-focusing in bulk damage experiments and present results of extensive studies we have conducted to verify several key features of the theory. We then discuss in more detail the relevance of the results in reference 2 to prior experiments.

## 2. Self-Focusing Theory

Marburger has addressed the problem of self-focusing for "pre-focused" beams in great detail (see reference 1, pp. 66-67). By "pre-focused" we mean a beam focused into the medium by an external lens. The approximate expression for the irradiance,  $I(z)$ , as a function of propagation distance ( $z$ ) in the nonlinear medium is given by:

$$I(z)/I(0) \cong \frac{1}{(1-z/R)^2 + (1-P/P_2)(z^2/k^2 a^4)} \quad (3)$$

$$\text{where } P_2 = \frac{3.77c \lambda^2}{32\pi^2 n_2} \quad (4)$$

$P$  = power in the beam

$k$  =  $2\pi/\lambda$

$a$  = beam waist at the entrance of the sample

$R$  = radius of curvature of the beam phase front due to the external lens

$\lambda$  = laser wavelength

$n_2$  = nonlinear refractive index

At the beam waist (focus) eq. (3) reduces to

$$I_{SF} \cong I_0 \left[ \frac{1}{1-P/P_2} \right] \quad (5)$$

where  $I_{SF}$  = peak irradiance in the presence of self-focusing

$I_0$  = peak irradiance without self-focusing

Equations (3) and (5) are approximations (sometimes referred to as the constant shape approximation) and are valid for

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 1 is a comparison of the approximation given by eq. (5) and the exact numerical solution given by Marburger (see reference 1, p. 66).

The factor of 3.77 in eqs. (1) and (4) 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 pre-focused and unfocused geometries. This means that for samples thicker than the Rayleigh range (as is the case for bulk laser-induced damage experiments) singular self-focus will occur within the sample and laser-induced damage will occur for 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 the results will be independent of self-focusing effects. The focal spot size needed will depend on the material  $P_2$  and the material breakdown threshold.

Equation (4) 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  $CS_2$ . 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 first step is to measure the  $n_2$  in a manner independent of the laser-induced breakdown measurements. Table 2 is a summary of such measurements for  $CS_2$  and other materials of interest using the technique described in reference [3]. Values obtained by other workers using various techniques are listed for comparison [7-10]. With one exception the agreement with other work is excellent. Note that as expected there is no dispersion in  $n_2$  for  $CS_2$  over the 532 to 1064 nm wavelength range and little or no dispersion in  $n_2$  for  $SiO_2$  and  $NaCl$ --contrary to the predictions of widely used theory for this phenomena [11].  $BK-7$  exhibits a negative dispersion in  $n_2$  over this wavelength range.

We set up a bulk breakdown experiment in  $CS_2$ , i.e., arranged the sample length to be much longer than the beam confocal parameter, and measured the breakdown power [12,13]. We then used eq. (4) to calculate  $n_2$  and compared this  $n_2$  to values obtained by beam distortion measurements [3,7]. The results of this comparison are shown in figure 1. In this experiment laser-induced breakdown is totally dominated by self-focusing and the breakdown power is independent of the focusing conditions [12]. Note the excellent agreement between the  $n_2$  determined from the breakdown measurements and eq. (4) and those determined by beam distortion measurements. This verifies that to the factor of 3.77 predicted by theory in reference 2 is correct to within the error bars shown.

Note the lack of dispersion in  $n_2$  in  $CS_2$  for the 532 nm and 1064 nm breakdown measurements. The  $n_2$  values are computed from  $P_2$  measurements using eq. (4). The beam distortion measurements confirm that there is no dispersion in  $n_2$  (the nonlinearity is due to nonresonant reorientation of the  $CS_2$  molecules). This implies that the  $\lambda^2$  dependence of  $P_2$  in eq. (4) is correct. This  $\lambda^2$  dependence is a well known consequence of self-focusing theory, however, to our knowledge this is the first direct confirmation of this wavelength dependence.

These results and those in reference 2 confirm is that  $P_2$  is the critical power of importance in laser-induced breakdown experiments. For input powers much smaller than  $P_2$  self-focusing effects will be minimal. As  $P$  approaches  $P_2$  self-focusing effects will dominate. For  $P < P_2/4$  one may use eq. (5) to correct for self-focusing effects and for  $P_2 > P > P_2/4$  one must use the full numerical solution to the nonlinear wave equation to calculate the irradiance at the focal position.

### 3. Relevance to Past Work

One conclusion of reference 2 was that past work in laser-induced breakdown needs to be re-examined in light of the new results regarding the role of self-focusing. This is what we attempt in this section. Much of the early experimental work [14-16] used the scaling law proposed by Zverev and Pashkov [17] in 1970. Fundamental to that technique is the assumption that the damage threshold irradiance for highly transparent materials is an intrinsic [18] property of the material and that any apparent dependence of the breakdown irradiance on focal spot radius is,  $w_0$ , in the material is due to self-focusing. The breakdown power for a given material was then measured as a function of focal spot radius and the results were fit to the equation [15]:

$$\frac{1}{P_B} = \frac{2}{I_B \pi w_0^2} + \frac{1}{P_1} \quad (7)$$

where  $P_B$  is the power required for breakdown,  $I_B$  is the so-called intrinsic damage irradiance and  $P_1$  is the first critical power for self-focusing as defined by eq. (2). [Note that eq. (7) can be derived from eq. (5) if  $P_2$  is replaced by  $P_1$  and the assumptions regarding intrinsic damage are made [15]. Such plots of  $1/P_B$  as a function of  $1/\text{area}$  were assumed to yield  $I_B$  as a slope and  $1/P_1$  as the y-intercept. Figure 2 is an example of such a plot for two samples of NaCl from the same vendor (Harshaw Chemical Co.). Note that the two plots yield values of  $P_1$  that differ by about a factor of 4 and the  $I_B$  is approximately an order of magnitude larger than commonly measured for this material. A similar plot of the results for sample 6 at 1064 nm yields  $P_1=0.70$  MW. This result is unreasonable in that  $n_2$  for NaCl has little if any dispersion in the wavelength region and therefore  $P_1$  at 1064 nm should be about 4 times larger than that at 530 nm (not equal as this result implies).

The data shown in figure 2 were taken in recent measurements in our laboratory and are presented in this manner simply to illustrate the problems associated with using the procedure discussed above. The main problems with the procedure are the assumption that  $I_B$  is intrinsic and the use of  $P_1$  in eq. (7).

Our conclusion in reference 2 was that prior damage results be reexamined. Unfortunately, much of the work using the Zverev and Pashkov [17] scaling is not recoverable from the literature since the uncorrected thresholds are not reported and cannot be extracted due to insufficient information. However Fradin [14], in anticipation of some problem with the Zverev and Pashkov method did not scale the data reported in 1973 for self-focusing. Smith et al. [16] in 1977 also found some problem in using  $P_1$  to correct 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,  $\phi(P)$ , was proposed such that the equation for the beam intensification as a function of input power is given by

$$I = \frac{I_0}{[1 - (P/\phi(P)P_1)]} \quad (8)$$

Numerical values for  $\phi(P)$  were proposed such that  $\phi(P)=1$  for  $P \ll P_1$  and  $\phi(P)=3.7$  for  $P \rightarrow P_2$ . However, since the exact functional dependence of  $\phi(P)$  was unknown at the time the thresholds reported, reference 16 included the breakdown power and the uncorrected focal area so that future workers could reexamine the data in the light of new measurements.

The results of this work indicate that  $\phi(P)$  is a constant with a numerical value probably close to 3.77, the numerical value predicted by Marburger [1] for Gaussian beams. If  $\phi(P)$  exhibited the functional dependence predicted by Smith et al. [16] then our experimental data should have indicated the presence of strong self-focusing for input powers below  $P_1$ . For example, if we take  $\phi(P)=1$  and let our input power equal  $0.9 P_1$  then eq. (8) would predict a factor of 10 increase in the peak-on-axis irradiance. This increase in the peak-on-axis irradiance in the material would result in an easily detectable change in the far-field transmitted beam profile. The fact that beam distortions and a polarization dependence are only seen in our test materials for beam powers approaching  $P_2$  indicate that  $\phi(P)$  is independent of power.

We will now use eq. 8 with  $\phi(P)=3.77$  to reexamine the 532 nm breakdown data of Smith et al. [16]. In that work, the breakdown powers are all below  $P_1$  so the irradiance increase predicted by eq. (8) with  $\phi(P)=3.77$  should be valid. In Table 3 we reproduce the 532 nm breakdown thresholds reported in reference 16. Six materials were studied including fused quartz and NaCl. The second and third columns contain the damage threshold power and the uncorrected focal area. The fourth column contains the ratio of the breakdown power to the  $P_1$  critical power for the material. The  $P_1$  values were calculated using  $n_2$  values measured by techniques other than interferometry [15]. The fifth column contains the ratio of the breakdown power to the critical power  $P_2$  calculated using the interferometric measurements of  $n_2$  of Weber et al. [18]. We use the  $n_2$  values of Weber et al. [18] to calculate  $P_2$  rather than scaling column four by the factor of 3.77, since the measurements in reference 18 are more accurate and are conducted using picosecond pulses. Thus the  $n_2$  values should be a reflection of the electronic nonlinear polarizability only. In the sixth column we list the R. M. S. breakdown fields in MV/cm uncorrected for self-focusing. Column

seven contains the data corrected for self-focusing using  $P_1$  as the critical power. These are the breakdown thresholds for 532 nm light published in reference 16. The number in parentheses represents the percentage increase of the corrected threshold over the uncorrected threshold. Finally in column eight we list the R. M. S. breakdown threshold fields corrected for self-focusing using the ratio  $P/P_2$  in column five. Again the numbers in parenthesis represent the percentage increase over the uncorrected threshold.

In examining the data in column seven we see that the breakdown threshold fields increased as much as 50-60 percent using  $P_1$ . However, when the thresholds are properly corrected using the second critical power  $P_2$  we see on the average only a 5 percent increase. This is well within the  $\pm 15$  percent absolute uncertainty in the uncorrected breakdown fields reported by Smith *et al.* [16] at 532 nm. In addition, the percentage increases in the breakdown threshold field for  $\text{SiO}_2$  could well be smaller (assuming the fused quartz sample of Smith *et al.* [16] (Suprasil I) has the same  $n_2$  value at high irradiance as our own Corning 7940 fused quartz).

Smith *et al.* [16] also reported breakdown thresholds at 355 nm laser wavelength in the materials listed in Table 3. 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  $0.7 P_1$ . The  $P_1$  critical powers were calculated using the  $n_2$  values measured at 1064 nm under the assumption that the dispersion in  $n_2$  over this wavelength range was small [16]. In an attempt to correct their data for the presence of self-focusing they scaled their breakdown threshold irradiance levels in  $\text{KH}_2\text{PO}_4$ , NaF, and LiF using eq. (8) and  $\phi(P)=2.5 \rightarrow 3.7$ . The fused quartz data was scaled using the same equation and  $\phi(P)=2.0 \rightarrow 3.0$ .

While they were on the right track, meaningful comparison of the 355 nm data with results at other wavelengths is difficult due to the very poor spatial quality of the 355 nm beam used in the measurements. The uncertainty in the 355 nm focal area and the uncertainty in the energy distribution within the focal area lead Smith *et al.* [16] to assign a factor of two range for the scaled breakdown threshold fields at this wavelength. The actual breakdown threshold fields may or may not be within this range. A further complicating factor for interpreting Smith *et al.* [16] 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 in BK-7 at 532 nm and those of White *et al.* [19] in BK-10 at 355 nm the  $n_2$  values for these materials may well be substantially lower at the third harmonic wavelength when compared to 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. As such we will not attempt at this time to reexamine the 355 nm thresholds.

In our reevaluation of the breakdown results of other workers we have concentrated on the work of Smith *et al.* [16] primarily 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 workers who scaled their breakdown thresholds for the presumed presence of self-focusing, Smith *et al.* [16] is the only work that contains sufficient data to deconvolve the true breakdown thresholds. Other workers merely reported the scaled breakdown threshold irradiance levels without including the focal spot radii used in the measurements [14,15,17]. This makes it impossible to recalculate the breakdown thresholds.

Several other breakdown studies have been conducted in these materials in which no self-focusing corrections were made. For example, Manenkov [20] 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 [20] work. Therefore, we will examine both cases. First, we must include the effects of electrostriction in our value for  $n_2$  since this mechanism is believed to play a strong role in the alkali halides for nanosecond pulse durations. We will use the value of  $n_2=4.0 \times 10^{-13}$  esu calculated from nanosecond three wave mixing experiments [21]. With this in mind we find that  $P_B$  in NaCl is  $0.5 P_2$  at 532 nm and  $P_B$  is  $0.14 P_2$  at 1064 nm if we use the larger of the two focal radii cited in reference 20. If we use the smaller focal radius we find that  $P_B$  for NaCl is  $0.07 P_2$  at 532 nm and  $0.02 P_2$  for 1064 nm. Thus self-focusing effects in Manenkov's [20] work for NaCl are negligible except for the combination of largest spot size and shortest wavelength.

In a similar nanosecond study, Merkle *et al.* [22] reported single shot damage thresholds for Corning 7940 fused quartz for laser wavelengths ranging from 1064 nm to 355 nm. In fused quartz electrostriction has been shown to play a small role in self-focusing effects for pulse durations

around 30 nsec [23]. Therefore, we will use the  $n_2$  value of  $0.95 \times 10^{-13}$  esu reported by Feldman *et al.* [23] for nanosecond pulse durations which includes contributions from electrostriction, thermal and electronic effects. We find that the breakdown powers,  $P_B$ , reported by Merkle *et al.* [22] for Corning 7940 are  $< 0.08 P_2$  at 1064 nm and equal to  $0.07 P_2$  at 532 nm. Therefore, self-focusing effects in the work of Merkle *et al.* [22] are negligible.

We have also reexamined [10] our own results published in references 5 and 6. In that work 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 distortions) were not conducted for each experimental condition used. In addition, there is a limit in the sensitivity of these techniques in detecting the presence of self-focusing, e.g., the time integrated beam distortion measurements can only detect a quarter-wave peak distortion. For the most part, little or no adjustment of the originally published numbers is needed. For the ultra-short pulse data (pulsewidth less than 5 psec) and largest spot sizes (14 micron  $1/e^2$  half-width) for  $\text{SiO}_2$  at 1054 nm adjustments as high as a factor of two were needed. However, for most of the  $\text{SiO}_2$  data only small adjustments were required. The corrected data showed essentially the same general dependences on pulsewidth and wavelength reported in references 5 and 6. For  $\text{SiO}_2$  and NaCl the pulsewidth dependence was consistent with the predictions of electron avalanche breakdown theory, while the wavelength dependence was not. The conclusion that bulk damage in these materials at 532 nm and 1064 nm is due to multiphoton initiated, electron avalanche breakdown is consistent with the corrected data. Even with self-focusing absent, a spot size dependence remains and sample to sample differences are observed. The latter results indicate the laser-induced breakdown is an extrinsic effect initiated by material defects.

#### 4. Summary

The key conclusions of this paper and reference 2 is that  $P_2$  is the critical power of importance in laser-induced damage experiments. Equation (5) can be used to correct for self-focusing if  $P < 0.25 P_2$ , otherwise the full numerical solution to the nonlinear wave equation must be used. Beam distortion and polarization dependence of laser-induced breakdown can be used to avoid experimental conditions for which self-focusing dominates the results.

The data reviewed in the last section of this paper is but a small sample of the laser-induced breakdown measurements available in the literature. We have examined a few specific examples for the purpose of illustrating the importance of using  $P_2$  instead of  $P_1$  in scaling breakdown results for self-focusing effects. It is important to note in Marburger's landmark review paper [1] he noted that Akhmanov's [24] constant shape approximation (which uses  $P_1$  and is the basis of the Zverev and Pashkov procedure) gives correct results only if  $P_1$  is replaced with  $P_2$  and  $P < P_2/4$ . Our work simply confirms this early theoretical conclusion.

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Table 1. Comparison of Constant Shape Approximation to Numerical Solutions for Focused Gaussian Beams

$P/P_2$	$(I/I_0)$ Constant Shape Approximation	$(I/I_0)$ Numerical Solution	Numerical Solution/ Constant Shape Approximation
0.27	1.37	1.30	0.95
0.60	2.50	2.08	0.83
0.80	5.00	3.94	0.79
0.90	10.00	7.15	0.72
0.95	20.00	16.45	0.82
0.96	25.00	28.60	1.14
0.97	33.30	63.40	1.92
0.98	50.00	100.00	2.00
0.99	100.00	192.00	1.92

Table 2.  $n_2$  Measurements in Liquids and Solids

Material	Wavelength ( $\mu\text{m}$ )	$n_2$	$n_2$
		( $\times 10^{-13}$ esu) This Work	( $\times 10^{-13}$ esu) Other Workers
CS <sub>2</sub>	1.06	128 $\pm$ 10	125 $\pm$ 30 <sup>a</sup>
	0.53	123 $\pm$ 10	
NaCl	1.06	1.37 $\pm$ 0.15	1.22 $\pm$ 0.21 <sup>b</sup>
	0.53	1.38 $\pm$ 0.13	
SiO <sub>2</sub>	1.06	0.62 $\pm$ 0.03	0.95 $\pm$ 0.10 <sup>b</sup>
	0.53	0.60 $\pm$ 0.04	
BK-7	1.06	1.45 $\pm$ 0.15	
	0.53	1.01 $\pm$ 0.08	

<sup>a</sup> K. J. Witte, M. Galanti, and R. Volk, Opt. Commun. 34, 278-282 (1980); Time Integrated Interferometry at 1.32  $\mu\text{m}$ .

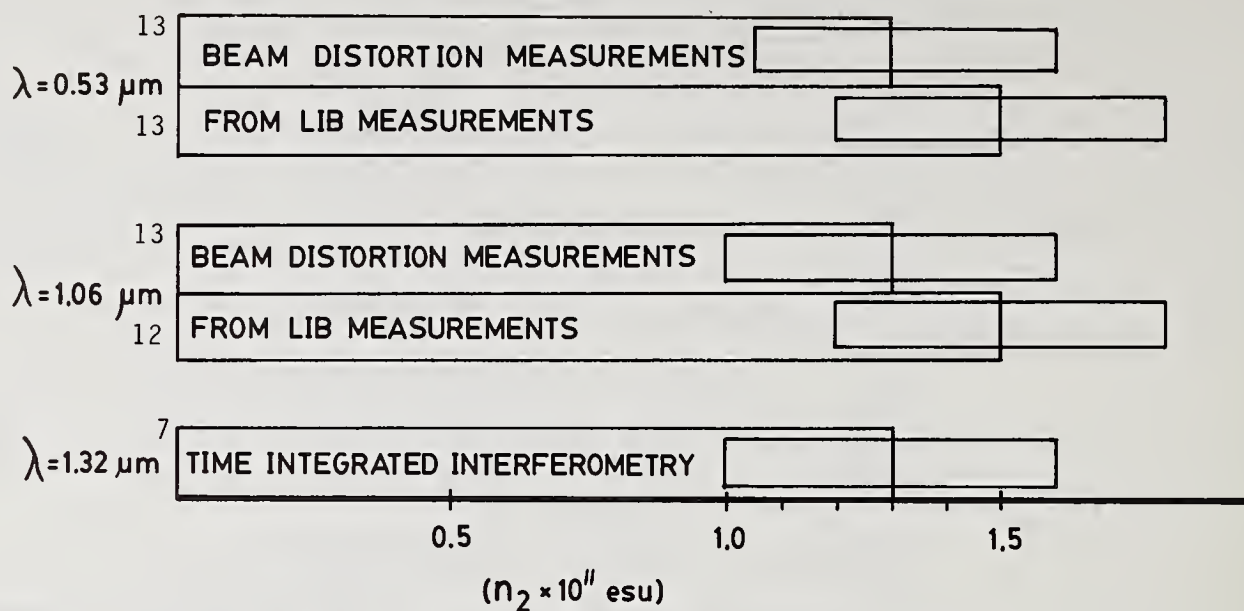
<sup>b</sup> M. J. Weber, D. Milam, and W. L. Smith, Opt. Eng. 17(5), 463-469 (1978); Time Resolved Interferometry at 1.06  $\mu\text{m}$ .

Table 3. 532 nm Breakdown Thresholds of Smith *et al.* [16]

Material	$P_B$ (KW)	$A$ ( $\mu\text{m}$ ) <sup>2</sup> (unscaled)	$P/P_1$	$P/P_2$	$E_0$ (MV/cm) (unscaled)	$E_{p_1}$ (scaled)	$E_{p_2}$ (scaled)
KH <sub>2</sub> PO <sub>4</sub>	151.0	16.0	0.57	0.15	15.3	23.4(53%)	16.6(8%)
SiO <sub>2</sub>	129.0	15.9	0.46	0.12	14.5	19.0(31%)	15.4(6%)
NaCl	38.4	15.1	0.60	0.05	7.9	12.4(57%)	8.1(3%)
CaF <sub>2</sub>	146.0	15.9	0.62	0.09	15.5	25.2(63%)	16.3(5%)
NaF	126.0	15.8	0.45	0.05	15.0	19.4(30%)	15.4(3%)
LiF	171.0	16.1	0.59	0.06	16.9	26.5(57%)	17.5(4%)

The last three columns contain the uncorrected breakdown fields and the values corrected for the presence of self-focusing. Column seven ( $E_{p_1}$ ) represents the data reported in reference 16. Column eight ( $E_{p_2}$ ) contains the data scaled using the correct critical power.





- <sup>13</sup> WILLIAMS, ET AL., OPTICS COMM., **50**, 256, 1984.  
<sup>12</sup> SOILEAU, ET AL., QUANT. ELECT. **QE-19**, 731, 1983.  
<sup>7</sup> WITTE, ET AL., OPTICS COMM. **34**, 278, 1980.

Figure 1.  $n_2$  from laser-induced breakdown (LIB) measurements. The LIB results were conducted by measuring the power required for breakdown for a sample of  $\text{CS}_2$  thick compared to the beam confocal parameter. Equation (2) was then used to calculate  $n_2$ .

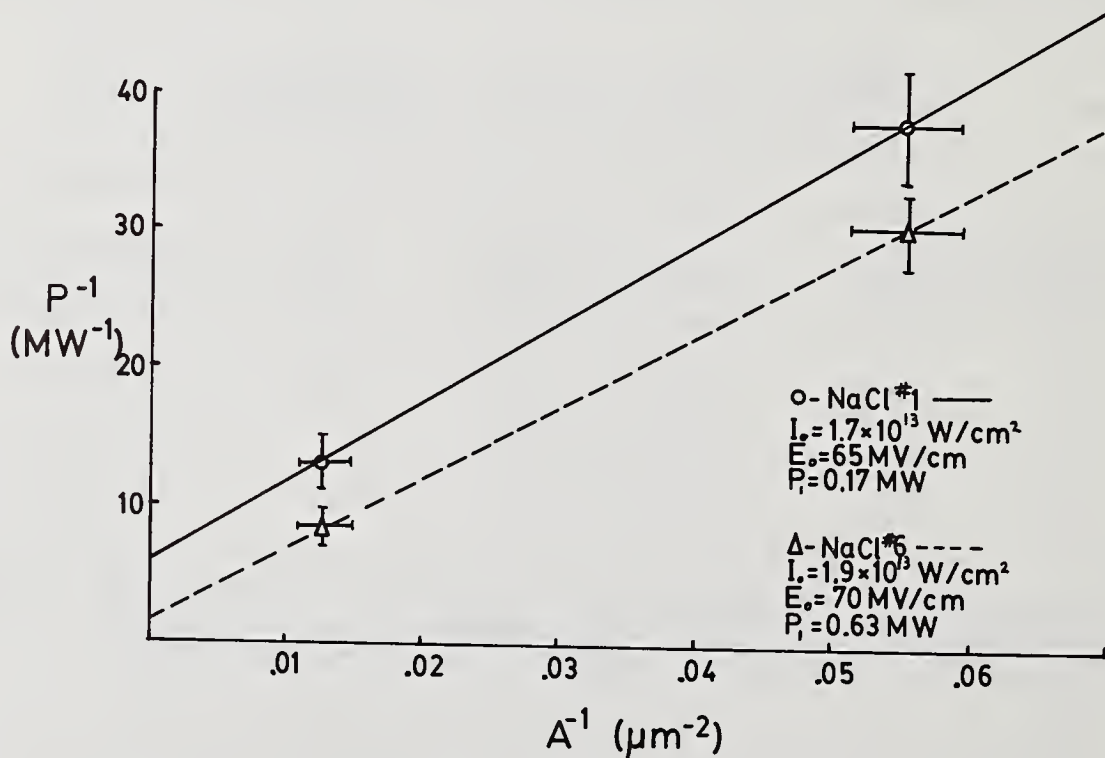


Figure 2.  $P^{-1}$  vs.  $(\text{Area})^{-1}$  for NaCl. These data are for linear polarized, 532 nm,  $35 \pm 10$  psec pulses. Note that these data are plotted in this manner to show that the procedure on which eq. (7) is based does not work! The  $I_B$  thusly determined are too large by an order of magnitude and the  $P_1$ 's for the two samples of the same material differ by a factor of 4.

*In response to the question as to theoretical predictions of whether  $n_2$  should go up with optical frequency, the author replied that the change in dispersion for picosec pulses is entirely because of electronic contributions and should go up with frequency, but only because of local field considerations, which would be expected to be minor. Experimentally it goes up significantly and the theory needs to be reexamined.*