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NATIONAL BUREAU OF STANDARDS REPORT

10 894

LASER DAMAGE IN MATERIALS

Semiannual Report I

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NATIONAL BUREAU OF STANDARDS

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LASER DAMAGE IN MATERIALS

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Solid State Materials Section
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LASER DAMAGE IN MATERIALS

I. Technical Report Summary

A. Technical Problem

The main objective of this program is the study of self-focusing in materials used in conjunction with high-energy laser systems. These studies are necessary because self-focusing is the main process which leads to bulk-intrinsic damage in laser materials. The damage appears as filamentary tracks in materials exposed to high-intensity laser radiation. Three mechanisms have been proposed to explain the effect: electrostriction, Kerr effect, and thermal self-focusing. By understanding the relative importance of these mechanisms, we hope to guide the development of materials with high self-focusing thresholds.

B. General Methodology

Laboratory experiments were conducted to study the process of self-focusing in three optical glasses: borosilicate crown glass, fused silica, and dense flint glass. The output of a Q-switched Nd:glass laser operating in the TEM_{oo} mode was focused into the samples. The laser beam was characterized fully as to pulse energy, pulse time evolution, beam profile, and reproducibility (see section II.B of Technical Report). Two different experiments were performed: (1) Self-focusing lengths, which are related to the damage track lengths, were measured as a function of laser beam peak power in order to test the current theory of self-focusing; (2) damage thresholds were measured for both linearly and circularly polarized radiation in order to distinguish between the proposed self-focusing mechanisms.

C. Technical Results

Measurements of self-focusing lengths as a function of peak power give good agreement with theory (see section II.C of Technical Report).

The ratio of damage thresholds for circular polarization to linear polarization is found to be greater than unity (see section II.D of Technical Report). The damage is assumed to result from self-focusing. These ratios indicate that, for the beam geometries used, both electrostriction and the Kerr effect are of comparable importance in producing self-focusing. Thermal self-focusing is considered to be negligible (see section II.A of Technical Report). The fractional contribution, α, of electrostriction to the nonlinear index n_2 is estimated (see Table I). The relative contribution of electrostriction decreases for larger focal spot sizes in agreement with the theory of electrostrictive self-focusing. The high damage threshold of fused silica is attributed to its relatively small Kerr effect.

D. Department of Defense Implications

The Department of Defense has a need for high-powered solid state laser systems. Thus it is important (1) to understand the processes which limit the output power of such systems, (2) to obtain data which suggest methods for increasing the power output of a given system, and (3) to verify theories which predict the performance limitations of such systems. We have shown that (1) electrostriction and the Kerr effect make comparable contributions to self-focusing for small beam diameters and for pulse widths of the order of 10 ns, but the contribution of electrostriction decreases for increasing beam diameters; (2) a higher damage threshold occurs with circularly polarized radiation than with linearly polarized radiation; (3) the current theory of self-focusing is essentially correct.

E. Implications for Further Research

Experiments will be continued on a larger number of materials including some laser glasses. Measurements will be extended to shorter pulse widths, from the present 25 ns to perhaps 2 ns. This will verify whether the 50% increase in self-focusing threshold for circular polarization over linear polarization predicted by the theory of the electronic Kerr effect can be realized experimentally. The present pulse width will be shortened by use of a laser-triggered spark gap and electro-optic shutter, which are currently under construction. Measurements are planned for wavelengths other than 1.06 μm to test the wavelength dependence of self-focusing. Frequency doubling will be tried first.

II. Technical Report*

A. Introduction

The mechanism responsible for the self-focusing of laser beams with pulse widths of ∿ 25 ns has not been uniquely identified. Three mechanisms have been proposed to explain the effect [1][1]: electrostriction [2-6], Kerr effect [2-4,7], and thermal self-focusing [8-10]. Up to the present time there has been insufficient data to judge the comparative importance of the proposed mechanisms. Self-focusing in liquids is caused by the Kerr effect due to the large contribution of freely rotatable molecules or ions to the nonlinear index coefficient n_2. Because solids lack freely rotatable molecules, some authors have thought that electrostriction [2-4] is the dominant mechanism. Duguay and Hanson have calculated the nonlinear index of refraction due to the Kerr effect $n_2(K)$ from measurements of induced birefringence in borosilicate crown glass using picosecond pulses [11,12]. They find that the value obtained is of sufficient magnitude to cause self-focusing and believe that the Kerr effect is the predominant mechanism for all pulse widths.

*The contents of this report were presented at the Fourth ASTM-NBS Symposium on Damage in Laser Materials, held in Boulder, Colorado, June 14 and 15, 1972.

[1] Figures in brackets indicate the literature references at the end of this paper.

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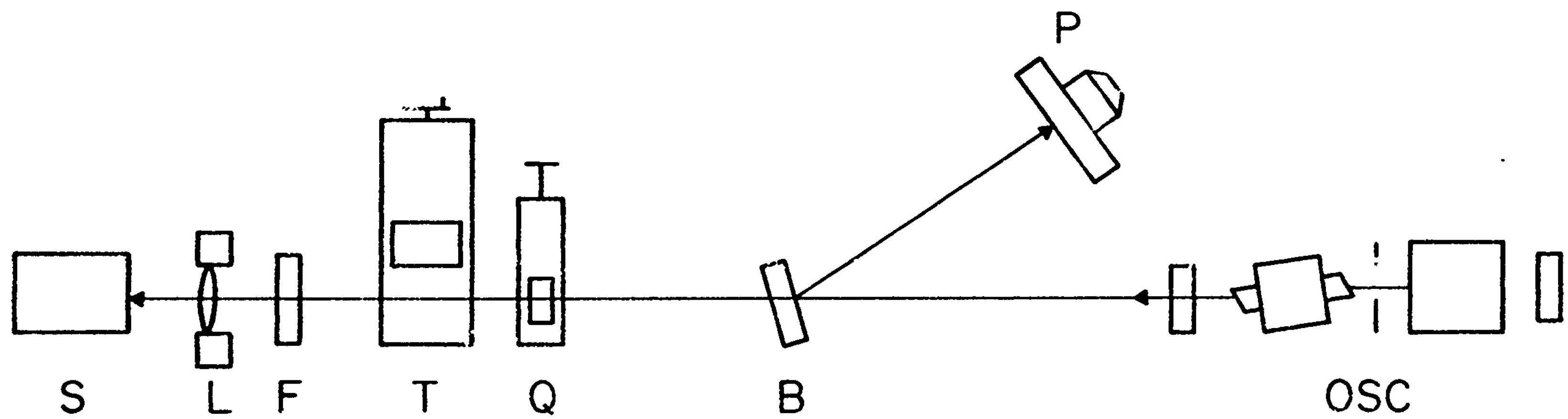

Fig. 1. Apparatus for damage testing: OSC = oscillator; B = beam-splitter; P = high speed photodiode; Q = quarter-wave plate for circular polarization, zero or one-half-wave plate for linear polarization; T = thermopile energy meter movable into the beam; F = attenuation filters; L = focusing lens; S = sample.

E. L. Kerr [6] has developed the theory of electrostrictive self-focusing to explain the data of Steinberg [13], who used 55-ns pulses from a ruby laser to measure damage thresholds in several optical glasses. Kerr states that electrostriction is the principal self-focusing mechanism for pulse widths $\gtrsim 10$ ns. For pulses less than 10 ns, on the other hand, he shows that the electrostrictive mechanism has insufficient time to take effect.

For the glasses which we are considering, thermal self-focusing is neglected. Quelle [10] in a sample calculation has shown that the threshold for thermal self-focusing would be two orders of magnitude greater than for electrostriction.

We have made self-focusing measurements in borosilicate crown glass (BSC 517), fused silica, and dense flint glass (SFS 5). We measured self-focusing lengths as a function of beam power [14] and compared them with the theory of Dawes and Marburger [15] which assumes an instantaneous self-focusing mechanism. Additional measurements showed that the damage threshold is higher for circular polarization than for linear polarization. From these measurements we estimated the relative importance of the proposed mechanisms to self-focusing. In liquids, similar measurements helped to confirm that the Kerr effect is the predominant mechanism [16,17].

B. Experimental Procedure

The experimental apparatus is shown in figure 1. The oscillator consists of a Nd:glass rod pumped by a helical flash lamp. It is Q-switched by means of a KD*P Pockels cell. There is an aperture in the cavity 2.44 mm

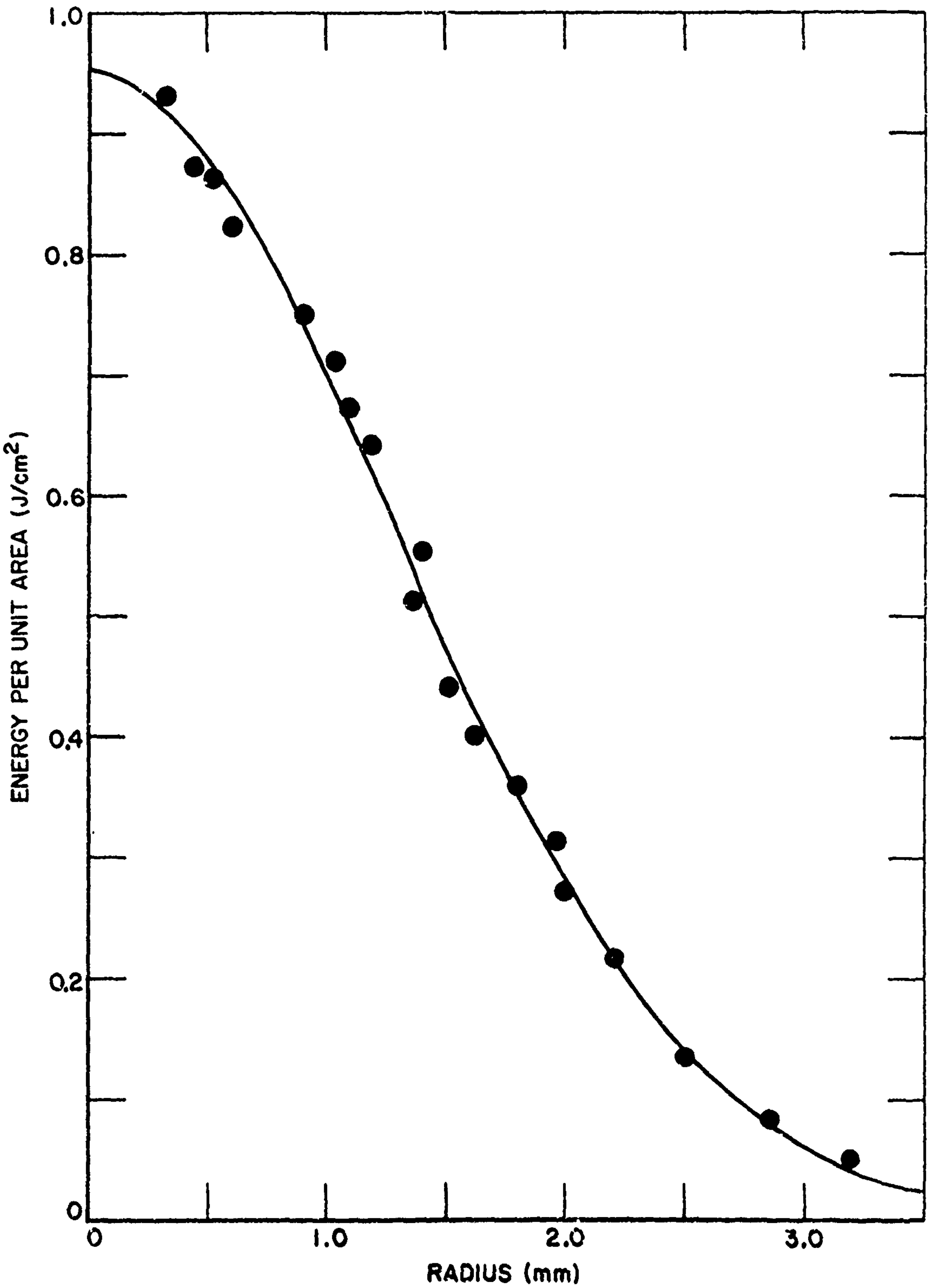

Fig. 2. Beam Profile. Energy per unit area as a function of distance from laser beam axis as measured at the focusing lens. The solid line is a Gaussian fit to the data.

in diameter for producing a beam in TEM_{00} mode. The output from the oscillator is a pulse whose width at half the maximum power is 25 ns as measured by a high-speed photodiode.

The beam profile of the laser beam at the focusing lens is obtained from burn patterns on developed unexposed black (Polaroid[2]) film [14]. The output of the laser is maintained at a constant pulse energy but is attenuated from pulse to pulse by interposing calibrated attenuation filters between the oscillator and the film. The radius of the outermost burn ring for each pattern is plotted as a function of beam energy. The result is fitted to a Gaussian curve $H = H_o e^{-r^2/a^2}$. The constant H_o is obtained in units of J/cm^2 from

$$E = 2\pi H_o \int_0^\infty \exp(-r^2/a^2) r \, dr, \qquad (1)$$

where E is the total pulse energy (99 mJ in this case). Figure 2 shows a plot of the beam profile together with the Gaussian which is fitted to the data by eye.

The energy output at a given pumping level was constant to within $\pm$ 2% as measured by the thermopile. During the damage tests each shot was monitored by the photodiode. The energy of the beam was varied in 20% increments with calibrated attenuation filters. Finer increments of energy were obtained by varying the oscillator pump energy.

[2]Certain commercial instruments and materials are identified in this report in order to specify adequately the experimental procedure. In no case does such identification imply recommendation or endorsement by the National Bureau of Standards, nor does it imply that the instrument or material identified is necessarily the best available for the purpose.

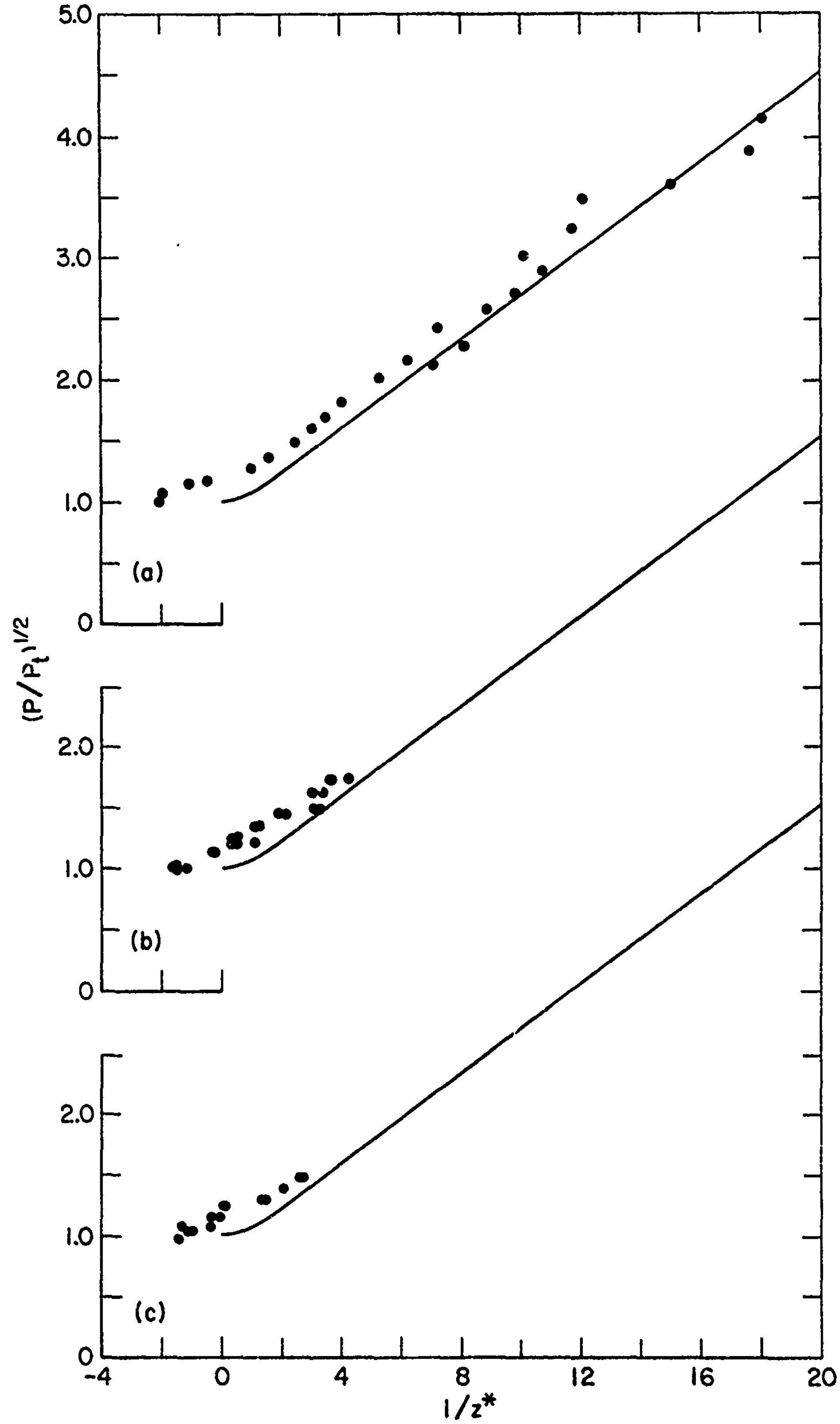

Fig. 3. Self-focusing length as a function of the square root of normalized beam power. A lens of 181-mm focal length was used to obtain the data. Samples used: (a) dense flint glass, (b) borosilicate crown glass, (c) fused silica. The solid curves are from the work of Dawes and Marburger [15].

C. Self-Focusing Length Measurements

Recent experiments [18-21] have shown that the filamentary damage observed when solids are exposed to high-intensity laser radiation is due to self-focusing of the laser beam and that the self-focused spot moves toward the laser source as the pulse power rises. Dawes and Marburger [15] have made a detailed computer calculation which predicts the self-focusing length as a function of beam power assuming that the nonlinear index coefficient n_2 responds instantaneously to the laser pulse shape.

We have produced damage filaments in optical glasses by focusing the laser beam at different power levels P into samples using a 181-mm focal length lens. We have measured a length z_f from the upstream end of the damage track to the sample entrance face and then have calculated the self-focusing length z from the formula [22]

$$z^{-1} = z_f^{-1} - R^{-1} \tag{2}$$

where R is the radius of curvature of the laser wavefront at the sample entrance face. R is calculated from the measured focal spot size of the lens and the propagation characteristics of Gaussian beams. The critical power for self-focusing P_t is taken to be the lowest power which produced damage in the sample. The normalized inverse self-focusing length $(z^*)^{-1}$ is plotted as a function of $(P/P_t)^{1/2}$ in figure 3 together with the theoretically calculated curve of Dawes and Marburger (our P_t is equivalent to their P_2), where $z^* = z/2ka^2$ and $k = 2\pi n_o/\lambda$; n_o is the refractive index of the medium; λ is the wavelength of the laser beam in air; a is the 1/e intensity point at the sample face. There is good qualitative agreement between the data and the theory. The deviations are attributed to electrostrictive self-focusing. Because of the low threshold in the SFS 5, we can compare experiment with theory over a large range of values. The fact that the slope of the data matches the experimental slope confirms that the laser beam had a Gaussian profile. We believe the data demonstrate that the theory provides a good description of self-focusing in solids.

D. Threshold for Linear and Circular Polarization

1. Experimental

Damage threshold measurements for linearly and circularly polarized light were made by focusing the laser beam into the samples using lenses of 76 mm and 181 mm focal length. The oscillator output was linearly polarized in the horizontal plane with the ratio of horizontal to vertical polarization being greater than 200:1. Circularly polarized light was produced by inserting a quarter-wave plate into the beam. The quarter-wave plate consisted of a plate of fused silica under uniaxial compression [23]. The linear polarization damage measurements were made by relaxing the stress in the plate. Measurements were also made with the plate stressed to a half-wave retardation. The results were consistent with the unstressed plate measurements.

Figures 4 and 5 show the damage threshold data. The solid points represent pulses that produced damage; the open points represent pulses that did not produce damage. The scatter in the data is due to

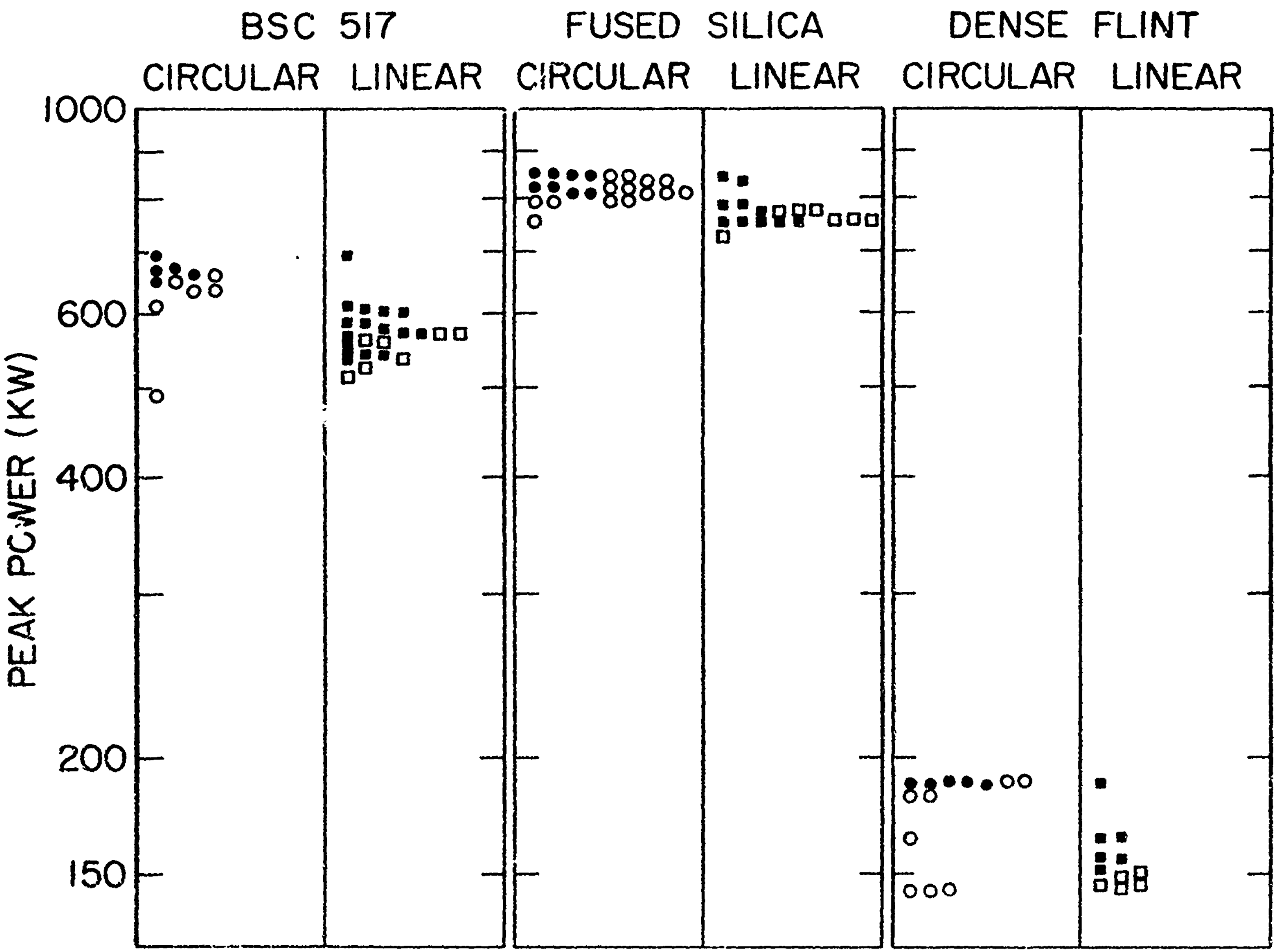

Fig. 4. Damage threshold measurements in borosilicate crown glass (BSC 517), fused silica, and dense flint glass (SFS 5) for linearly and circularly polarized radiation; wavelength = 1.06 μm; pulse width = 25 ns. The solid points represent shots which produced damage; the open points represent shots which did not produce damage. The data are obtained by using a 76-mm focal length lens.

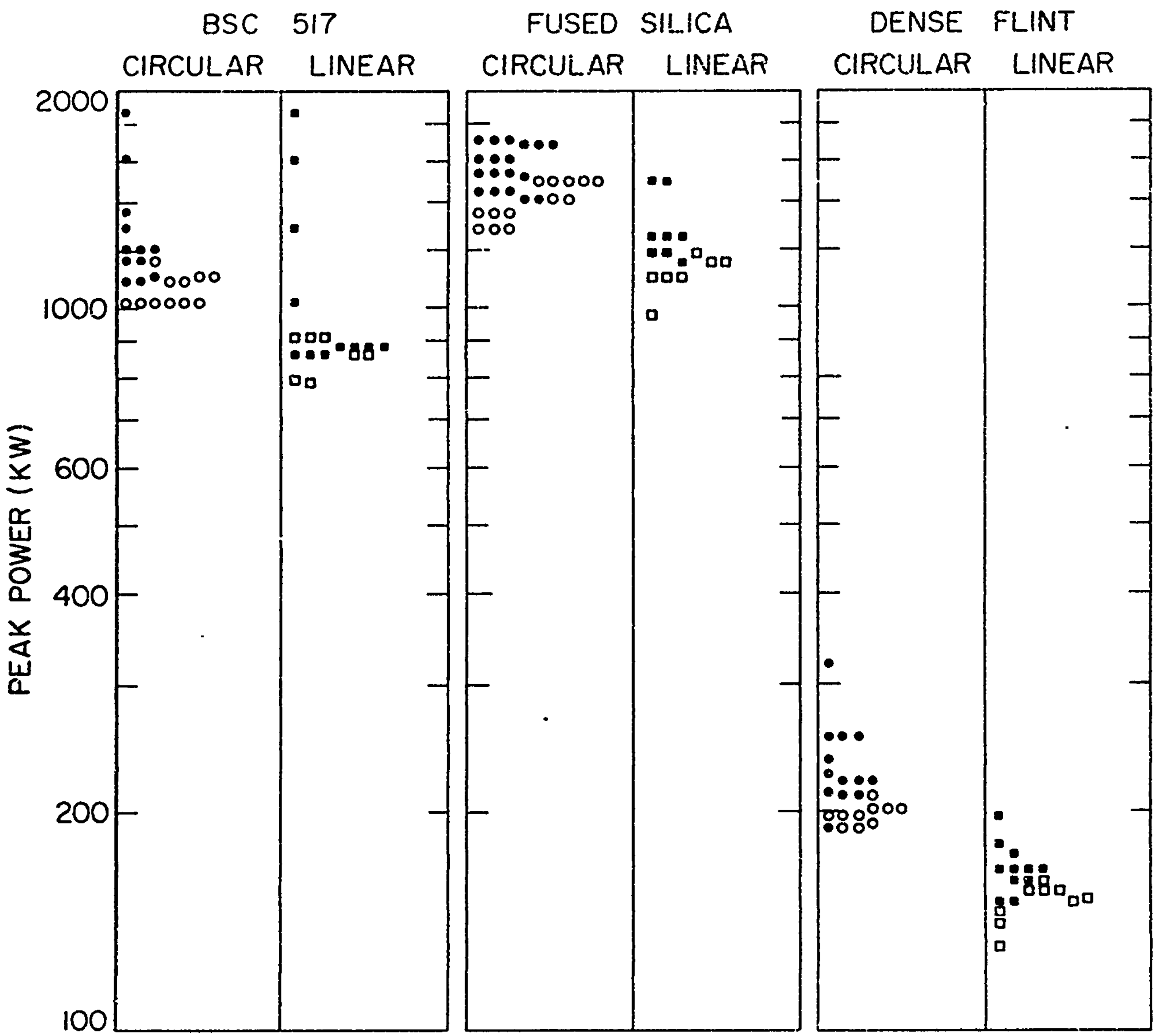

Fig. 5. Damage threshold measurements in borosilicate crown glass (BSC 517), fused silica, and dense flint glass (SFS 5) for linearly and circularly polarized radiation; wavelength = 1.06 μm; pulse width = 25 ns. The solid points represent shots which produced damage; the open points represent shots which did not produce damage. The data are obtained by using a 181-mm focal length lens.

sample variability, laser energy variations, and the statistical nature of the actual damage process [20]. In all cases, the threshold for circularly polarized light P_t' is higher than for linearly polarized light, P_t. (Unprimed symbols refer to linear polarization and primed symbols to circular polarization.) Table 1 lists the ratios P_t'/P_t. The error limits represent the full scatter in the data.

2. Relation of Experiment to Theory

The mechanisms of electrostriction and the Kerr effect are associated with the quadratic dependence of refractive index on electric field E: $n = n_o + n_2 E^2$; $n_2 = n_2(es) + n_2(K)$. [es ≡ electrostriction, K ≡ Kerr effect.] The self-focusing threshold P_t is inversely proportional to n_2. The relative contributions of the above mechanisms to the self-focusing can be calculated from the experimental data if we make several assumptions: (1) The damage is assumed to result from self-focusing; (2) thermal self-focusing can be ignored; (3) the ratios $n_2(es)/n_2'(es)$ and $n_2(K)/n_2'(K)$ can be calculated.

In calculating $n_2(es)/n_2'(es)$, we assume the material responds to a D.C. field obtained from a time average of the optical electrical field tensor E_iE_j. It is assumed that the E field is uniform within a cylinder and zero outside. The result will b the same on the axis of a Gaussian beam. Detailed dynamical calculations are not expected to change the ratio $n_2(es)/n_2'(es)$ even though they would affect the absolute index changes. Steady-state conditions are assumed.

Table 1. Parameters for calculating electrostriction and Kerr effect in optical glasses.

	BSC 517	Fused Silica	Dense Flint
n_o (1.06 μm)	1.507	1.450	1.733
q_{11} (10^{-13} cm^2/dyn)	[a]0.315	[b]0.418	[c]1.7
q_{12} (10^{-13} cm^2/dyn	[a]1.92	[b]2.71	[c]2.2
p_{11}	[a]0.115	[b]0.120	[c]0.21
p_{12}	[a]0.221	[b]0.270	[c]0.23
n_2(es) (10^{-13} esu)	0.62	0.81	2.5
n_2'(es) (10^{-13} esu)	0.56	0.72	2.5
$n_2(es)/n_2'(es)$ (theory)	1.11	1.13	1.00
$n_2(K)/n_2'(K)$ (theory)	1.50	1.50	1.50
$P_t'/P_t = n_2/n_2'$ (experimental)			
Reference d	1.18 ± 0.07	1.09 ± 0.09	1.25 ± 0.08
Reference e	1.26 ± 0.12	1.24 ± 0.11	1.32 ± 0.07
α			
Reference d	0.8 ± 0.2	1.15 ± 0.35	0.40 ± 0.13
Reference e	0.6 ± 0.3	0.7 ± 0.3	0.27 ± 0.13

[a]R. M. Waxler and C. E. Weir, J. Res. Natl. Bur. Std. (U.S.) 69A, 325 (1965), λ = 587.6 nm.

[b]W. Primak and D. Post, J. Appl. Phys. 30, 779 (1959), λ = 546.1 nm.

[c]This work, λ = 632.8 nm.

[d]Data obtained with 76-mm focal length lens. Error limits represent full scatter in the data.

[e]Data obtained with 181-mm focal length lens. Error limits represent full scatter in the data.

In the presence of an intense uniform electric field all solids undergo a strain [24]

$$\epsilon_{k\ell} = \gamma_{ijk\ell} E_i E_j . \tag{3}$$

From thermodynamics we can show that

$$\gamma_{ijk\ell} = - \kappa_{im} \kappa_{jn} q_{mnk\ell}/4\pi , \tag{4}$$

where κ_{ij} is the dielectric tensor and $q_{ijk\ell}$ is the stress-optical coefficient. If we assume a freely expanding cylinder, we obtain the expressions

$$n_2(es) = n_o^7 (p_{11}q_{11} + 2p_{12}q_{12})/8\pi \tag{5}$$

$$n_2'(es) = n_o^7 (p_{11}q_{11} + 3p_{12}q_{12} + p_{12}q_{11} + p_{11}q_{12})/16\pi , \tag{6}$$

where the p_{ij} are the elasto-optic coefficients; the Voigt notation is used. Values for the above constants are given in Table 1. Although the numbers quoted were obtained at visible wavelengths, we do not expect them to differ significantly at 1.06 μm.

For the Kerr effect we assume only an electronic contribution [11,12,25-27]. Recent experimental work suggests this is indeed the case [28]. For an isotropic material the nonlinear polarization is [29]

$$\vec{P}^{NL} = 3C_{1122} (\vec{E}\cdot\vec{E})\vec{E} . \tag{7}$$

If we insert the field for a linearly polarized wave $\vec{E} = [E_o \cos(\omega t-kz), 0,0]$ and a circularly polarized wave $\vec{E} = [E_o \cos(\omega t-kz), E_o \sin(\omega t-kz), 0]/\sqrt{2}]$ into Eq. (7) and calculate the Fourier component of $\vec{P}^{NL}$ at frequency ω, we obtain a ratio of 1.5 for $n_2(K)/n_2'(K)$.

It is now possible to estimate the fractional contribution of electrostriction, $\alpha = n_2(es)/n_2$, and the Kerr effect, $(1-\alpha) = n_2(K)/n_2$, to the nonlinear index coefficient. Calculated values for α are summarized in Table 1.

One cannot attribute any significance to one value of α being greater than unity for SiO_2. This value is obtained with a lens of short focal length. Because the self-focusing threshold for fused silica is high, it is probable that only a slight amount of self-focusing took place and the threshold measured is essentially the intrinsic damage threshold for which one might expect no difference between circular and linear polarization.

The data indicate that the Kerr effect is larger in BSC 517 than in SiO_2. This conclusion would explain why fused silica has a higher damage threshold than BSC 517 since, on the basis of electrostriction alone, BSC 517 would have the higher damage threshold. In dense flint glass the Kerr effect appears to make a greater contribution than electrostriction; the observed difference in damage threshold between circular and linear polarization must be ascribed to the Kerr effect since no difference is expected from electrostriction alone.

In all cases the value of α is larger for the longer focal length data. This is reasonable since electrostrictive effects are expected to decrease with increasing focal spot size while the Kerr effect is expected to remain unchanged. It is interesting to note that electrostriction makes a relatively smaller contribution in SFS 5 than in the other glasses. This is to be expected since electrostriction depends on the parameter

$X = a_o/v\tau$ where τ is the pulse width and v the velocity of sound in the medium [6]. As X increases, electrostrictive effects decrease. For SFS 5, v = 3000 m/sec, while for SiC_2 and BSC 517, v = 6000 m/sec. Thus, X is twice as large for SFS 5 as it is for the other glasses.

Figure 6 compares our data with the data of Steinberg [13] and E. L. Kerr's theory of electrostrictive self-focusing [6]. The figures are obtained from Kerr's work. The X's represent values we measured. In order to compare the data, it was necessary to scale the power with λ^2 and the beam radius with $1/\tau$. We find relatively good agreement for the fused silica and borosilicate crown data with that of Steinberg. The data for SFS 5 do not agree, however. Our data seem reasonable in view of the previous results. In SFS 5 the relative contribution of electrostriction is small, and hence we expect a small change in damage threshold for the different lenses used. In fused silica and BSC 517, where electrostriction appears to be more important, we observe a definite increase in damage threshold with a longer focal length lens.

These data suggest that the Kerr effect increases with refractive index. A large refractive index implies large charge displacements, and large charge displacements are more likely to lead to a nonlinear response to the electric field, and therefore, to a large Kerr effect. It is known that flint glasses exhibit increasing polarizability and refractive index with increasing lead content [30]. The data obtained are consistent with axis rotation measurements of elliptically polarized light [28].

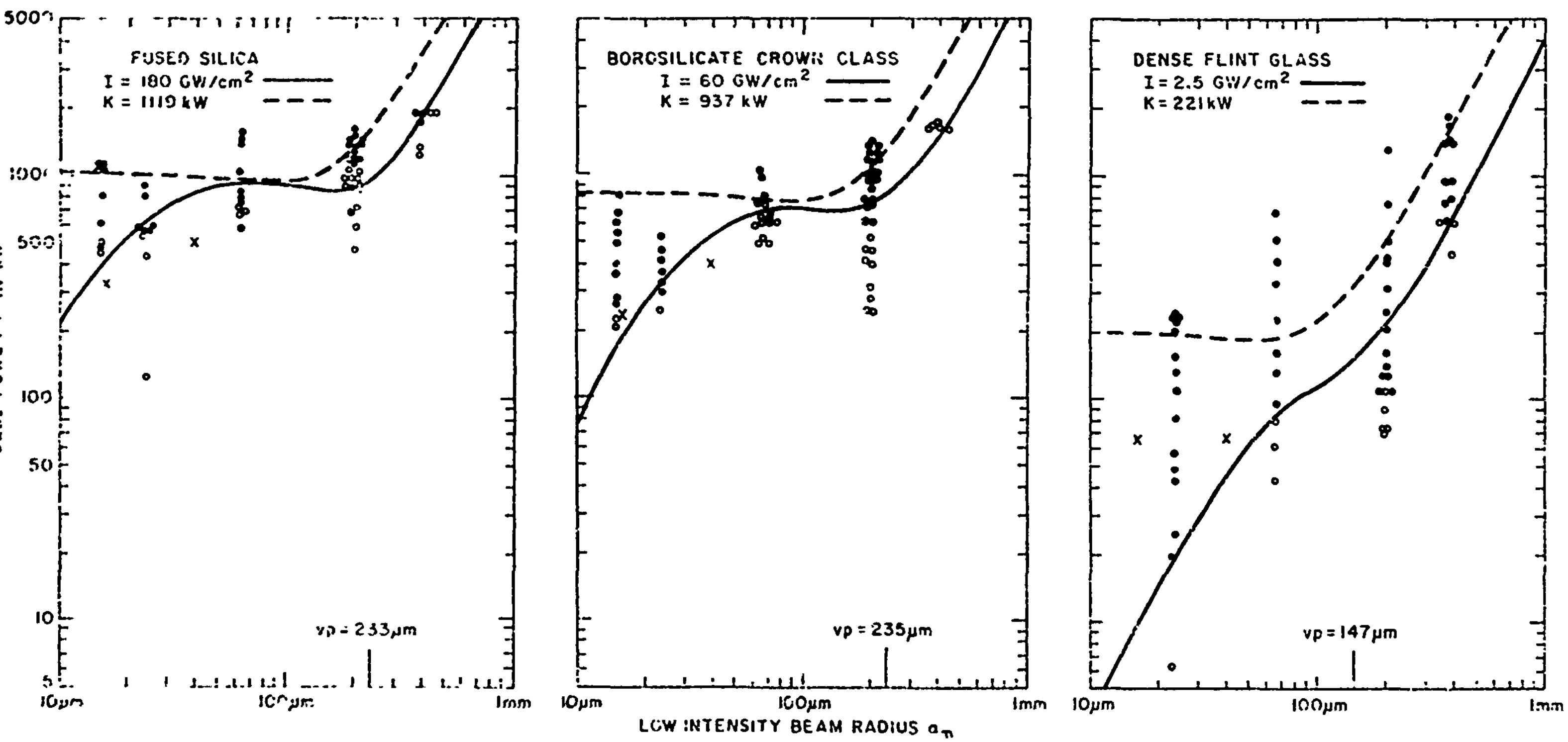

Fig. 6. Track formation thresholds in borosilicate crown glass, fused silica, and dense flint glass from the work of E. L. Kerr [6] and G. N. Steinberg [13]. The data from this work are depicted by X. It is necessary to scale our data as the wavelength and pulse width used differ from Steinberg's.

E. Conclusions

Our self-focusing length measurements show that the theory of Dawes and Marburger provides a good description of self-focusing.

From threshold measurements using linearly and circularly polarized radiation we conclude that the Kerr effect and electrostriction are of comparable magnitude for the beam geometry used.

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