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Laser Damage in Materials

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National Bureau of Standards
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Abstract

This report summarizes the study of damage and self-focusing in materials used in Q-switch solid-state laser systems. In borosilicate crown glass, fused silica, dense flint glass, and yttrium aluminum garnet, self-focusing appears to be the main cause of damage. An analysis of damage threshold measurements with linearly polarized radiation and circularly polarized radiation suggests that the Kerr effect is the dominant self-focusing mechanism with a significant contribution to self-focusing from the thermal effect. The electrostrictive effect is negligible. The damage threshold in Nd:doped laser glasses appears to be intrinsic. In all the above materials, the damage threshold for circular polarization is greater than the damage threshold for linear polarization. In lithium niobate, calcite, potassium dihydrogen phosphate, and deuterated dihydrogen phosphate, damage at the lowest levels is caused by inclusions. Bulk and surface damage thresholds in Nd:doped thoria:yttrium oxide ceramic are obtained relative to bulk damage thresholds in several optical materials. Relationships under different geometric boundary conditions are also derived for solid materials between the stress-optic coefficients and the electrostrictive coefficients.

LASER DAMAGE IN MATERIALS

1. Technical Report Summary

1.1 Technical Problem

        The main objective of this program is the measurement of damage
thresholds and the determination of mechanisms associated with self-
focusing in materials used in high-energy pulsed solid-state laser systems.
The materials we have examined are used in the fabrication of optical com-
ponents, such as lenses, beamsplitters and mirrors, laser rods, modulators,
Q-switches, and polarizers.

        Self-focusing is the main process which leads to intrinsic
bulk 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. The study of self-focusing
can be obscured, however, if extrinsic damage processes, such as inclusion
damage, take place.

1.2 General Methodology

        Laboratory experiments were conducted to determine the mechanisms
of bulk damage in the following materials:

        borosilicate crown glass (BSC 517)
        fused silica
        dense flint glass (SF 55)
        yttrium aluminum garnet (YAG)
        five commercial Nd:doped laser glasses
        calcite ($CaCO_3$)
        potassium dihydrogen phosphate (KDP)
        deuterated potassium dihydrogen phosphate (KD*P)
        lithium niobate ($LiNbO_3$)
        neodymium-doped thoria:yttrium oxide ceramic.

The output of a Q-switched Nd:glass oscillator operating in the TEM_{oo} mode
was focused into the samples. The laser beam was characterized as to
pulse width, pulse time evolution, beam profile, and reproducibility.

Damage sites in the samples were examined to ascertain whether inclusion damage or self-focusing induced damage was the limiting damage process. In samples where self-focusing was the principal mechanism of damage, we performed two different experiments: (1) Damage thresholds were measured for both linearly and circularly polarized radiation; (2) damage track positions, which are related to self-focusing lengths, were measured as a function of laser beam peak power. A theoretical analysis of the data was made by assuming that the experimental damage thresholds could be used to determine the self-focusing threshold. The contributions of the Kerr, electrostrictive, and thermal effects to the intensity dependent refractive index were theoretically analyzed. The measurement of damage thresholds for linearly and circularly polarized radiation provided the necessary and sufficient data from which to estimate the relative importance of each of the effects to self-focusing. The self-focusing length data were compared to the theoretically predicted values.

1.3 Technical Results

Self-focusing appears to be the principal cause of bulk damage in BSC 517, fused silica, SF 55, YAG, and Nd:doped laser glass. In all these materials, we find that the damage threshold for circularly polarized radiation is greater than the damage threshold for linearly polarized radiation. A lower bound to the nonlinear index n_2 and the contributions of the Kerr effect, electrostriction and the thermal effect to n_2 were obtained from a detailed analysis of the data in BSC 517, fused silica, SF 55, and YAG. For the pulse width (25 ns) and beam geometry used, the Kerr effect appears to be the largest contributor to self-focusing with a significant contribution from the thermal effect. Electrostriction is the smallest of the effects and it would become negligible for an unfocused beam. Reasonable values of the absorption coefficient are obtained from the estimated thermal effect. In the glasses tested, the Kerr effect appears to increase with increasing refractive index.

Using a focused geometry with a 181-mm focal length lens, we find that damage track position measurements in BSC 517, fused silica, SF 55, and YAG agree well with self-focusing lengths predicted by theory. On the other hand, the agreement with theory is poor in the Nd:glasses and in data obtained with a 362-mm focal length lens in SF 55. The discrepancy occurs because the position of a damage point on the filament does not necessarily coincide with a self-focusing point. No significant difference was found in the damage thresholds among the laser glasses.

We find in calcite, KDP, KD*P, and $LiNbO_3$ that inclusions are the limiting cause of damage. In $LiNbO_3$ we also find evidence for self-focusing, but this occurs at higher power levels.

Bulk damage measurements were made in a neodymium-doped thoria:yttrium oxide ceramic laser rod in cooperation with Charles Greskovich of the General Electric Corporate Research and Development Laboratory*. The damage threshold relative to several optical materials was tabulated.

We have obtained relationships between the stress-optic coefficients and the electrostriction coefficients for three geometric situations. Our derivation is based on earlier theoretical work. We find that the differing results of several authors correspond to solutions for different geometric boundary conditions.

1.4 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 output power of a given system, and (3) to verify theories which predict the performance of such systems. We have come to the following conclusions: (1) In materials for which self-focusing is the dominant mechanism leading to damage, such as glasses, sapphire, and YAG, the Kerr effect is the dominant self-focusing mechanism. The Kerr effect increases with increasing refractive index; therefore, for this type of materials, those having low refractive indices will have high self-focusing thresholds. (2) The thermal effect can be important for pulse widths $\geq$ 25 ns or for high repetition rate systems. (3) Inclusion damage limits the performance of many crystals used presently in modulators and Q-switches, such as calcite, KDP, KD*P, and $LiNbO_3$. In order to obtain higher damage thresholds, methods must be found to eliminate the inclusions. When this is done, self-focusing will then become the limiting process.

1.5 Implications for Further Research

The interests of the Department of Defense have been shifting to longer wavelengths in the infrared (10.6 μm for the CO_2 laser and 3.8 μm for the DF laser). Interest is also expected to shift toward the ultra-violet region even though there are still no important high-power lasers in this region. Over this wide wavelength range there is a lack of data on optical materials that could be used for the construction of optical components. These data include changes of refractive index with temperature and stress which are important because components subjected to high-intensity radiation can undergo a significant change of refractive index

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due to heat generated by low-level absorption processes. These changes in index are caused by variations of temperature and by stresses introduced by temperature gradients. Therefore, measurements of the refractive index and the change of refractive index with temperature and stress, in both the infrared and ultraviolet regions of the spectrum, are worthwhile. Measurements in the infrared of this nature are presently occurring at the National Bureau of Standards under a joint program with ARPA.

2. Technical Report

2.1 Introduction

The output of high-powered laser systems is limited by the damage occurring in components exposed to the intense electromagnetic radiation. An understanding of the mechanisms underlying the damage process is necessary if we are ever to fabricate, in a systematic manner, materials with high self-focusing thresholds. In the past several years, significant progress has been made in understanding these underlying mechanisms. For example, it has been shown that the phase shift due to Fresnel reflection is the mechanism responsible for the fact that the damage threshold of an entrance surface is greater than the damage threshold of an exit surface [1]. In addition, the surface damage threshold has been significantly increased by appropriate polishing techniques which eliminate damaging inclusions in the surface region [2,3]. Thus the intrinsic surface damage threshold is thought to be the same as the bulk damage threshold [3].

In this report we summarize the conclusions [4-6] reached during the course of our work over the past two years. We include summaries of our work described in previous reports together with minor modifications. In all cases the experimental work was done with a Nd:glass Q-switched laser operating in the TEM_{OO} mode with a temporal pulse width of 25 ns.

Our work has been concerned with determining the mechanisms underlying bulk damage. This work includes the study of self-focusing, which is the main process leading to bulk intrinsic damage in the components of Q-switched solid-state laser systems. We have estimated the relative importance of the Kerr effect, electrostriction, and the thermal effect to self-focusing in materials in which self-focusing is the principal bulk damage process [5,7,8]. In other materials we found that inclusions are the limiting cause of damage [6].

4.

2.2 Self-Focusing Studies in Optical Glasses and YAG

Damage in the form of filamentary tracks was observed when 1.06 μm laser radiation was focused into samples of BSC 517, fused silica, SF 55 and YAG. This filamentary damage indicated that self-focusing was the process leading to damage in these materials. We therefore set out to determine which of the proposed self-focusing mechanisms, electrostriction, the Kerr effect, or the thermal effect, was the dominant self-focusing mechanism. By knowing which mechanism predominates, one might then be able to tailor materials with high self-focusing thresholds.

We have estimated the relative contributions of the Kerr effect, electrostriction, and thermal effects to the self-focusing thresholds of BSC 517, fused silica, SF 55, and YAG from an analysis of damage threshold data for linearly polarized and circularly polarized radiation. In all cases, the damage threshold for circular polarization was higher than the damage threshold for linear polarization. For the beam geometry used, we conclude that the Kerr effect is the largest effect in all the samples tested. The thermal effect is also found to be significant, but the electrostrictive effect is quite small. Both of the latter effects are expected to be insignificant for an individual subnanosecond pulse; however, because of its integrating nature, thermal self-focusing can be important in high-repetition-rate laser systems.

The results we have obtained are in qualitative agreement with the work of others. Absorption coefficients calculated from the estimated thermal contribution to self-focusing appear to be reasonable for commercial optical materials.

The assumptions in the analysis tend to minimize the values we obtain for $n_2(K)$. In addition, the thermal contribution may be enhanced at the expense of $n_2(K)$. Nevertheless, these considerations do not weaken but strengthen our basic conclusion, that the Kerr effect is the dominant self-focusing mechanism. The numbers we calculate indicate that absorption coefficients of the magnitude present in commercial optical materials can make a significant contribution to self-focusing.

In this summary we include some modifications to the results in our earlier report. These modifications have appeared in the literature [7,8]. In the analysis, in addition to using the damage threshold data obtained with the 181-mm focal length lens, we also include data obtained with the 76-mm focal length lens. We then obtain the critical power for self-focusing

$$P_{cr} = (f_2^2 - f_1^2)/\{[f_2^2/P_t(2)] - [f_1^2/P_t(1)]\}, \tag{1}$$

where $P_t(1)$ and $P_t(2)$ are damage thresholds obtained with lenses of focal lengths f_1 and f_2, respectively. If we assume that $P_{cr} = P_1$ from the theory of Dawes and Marburger [9], we then obtain a lower bound to the nonlinear index n_2. The change of results for the optical glasses plus a change of laser beam calibration lead to a change in Table II of reference [5]. The revised table (Table I) is included herewith. Results for YAG are contained in references [5-7].

In analyzing the contribution of electrostriction to self-focusing, we made use of the following relationship between the stress-optic coefficients q and the electrostrictive coefficient γ:

$$\gamma_{ijkl} = - \kappa_{im} \kappa_{jn} q_{mnkl}/4\pi,$$

where κ is the dielectric tensor. A different result was obtained by Maradudin and Burstein [10]. We showed that the different results are due to different geometric boundary conditions, much as the polarization of a solid in an electric field will depend upon the shape of the solid [7].

2.3 Relationship Between Self-focusing Lengths and Damage, Including Damage in Nd:Doped Laser Glasses [4,5]

Assuming that a point of damage on a damage track corresponds to a point of self-focusing, we plotted self-focusing lengths as a function of peak beam power and compared the results with the theoretical curve of Dawes and Marburger [9]. We found surprisingly good agreement between theory and experiment in BSC 517, fused silica, SF 55, and YAG when we used the 181-mm focusing lens. There was gross disagreement with theory for data obtained in five Nd:doped laser glasses. In addition, there was disagreement when radiation was focused in SF 55 with a 362-mm focal length lens. We propose the following explanation for the discrepancy. For the data in which there is agreement with theory, the agreement is fortuitous in that there was an insignificant difference between a point of self-focus and a point of damage. This difference, in general, may be significant because the intensity will exceed the damage threshold upstream from a point of self-focus. The self-focus point is where the energy density is theoretically infinite. An extreme case occurs when damage takes place without self-focusing or with weak self-focusing. In this case there is no singularity in the energy density. We believe the damage in the laser glasses to be this latter case. Assuming self-focusing is negligible, we have calculated the damage threshold of the laser glasses. The results, which are given in Table II, are based on a focal spot size measured in air. The spot size is approximately one and one-half times the theoretical size of a focused beam with a Gaussian profile.

Table I. Contributions of Kerr, electrostrictive, and thermal effects to self-focusing thresholds[a]

	BSC 517	Fused Silica	Dense Flint SF 55
$P_t(1)$[b] (MW)	0.47 ± .06	0.65 ± .07	0.124 ± .015
$P_t(2)$[c] (MW)	0.75 ± .09	1.01 ± .12	0.128 ± .015
$P_t'(1)$[b] (MW)	0.55 ± .07	0.71 ± .08	0.153 ± .018
$P_t'(2)$ (MW)	0.94 ± .13	1.25 ± .15	0.170 ± .018
P_{cr} (MW)	0.86 ± .14	1.15 ± .18	0.129 ± .018
P_{cr}' (MW)	1.10 ± .20	1.49 ± .25	0.174 ± .022
n_2 (10^{-13} esu)	1.24 ± .20	0.93 ± .15	8.3 ± 1.2
n_2' (10^{-13} esu)	0.97 ± .18	0.72 ± .12	6.1 ± 0.8
n_2(ES) [10^{-13} esu]	0.13	0.16	0.68
n_2'(ES) [10^{-13} esu]	0.11	0.13	0.68
n_2(K) [10^{-13} esu]	0.80 ± .38	0.56 ± .27	6.4 ± 1.9
n_2(T)	0.29 ± .27	0.19 ± .19	1.1 ± 1.2
α (cm^{-1})	$(3 \pm 3) \times 10^{-3}$	$(9 \pm 9) \times 10^{-4}$	$(7 \pm 7) \times 10^{-3}$

[a]The errors occur from the scatter in the data and from the uncertainty in the power calibration.

[b]Obtained with 76-mm-focal-length lens [4]. The data have been corrected for thermopile calibration and for sample entrance face reflection.

[c]Obtained with 181-mm-focal-length lens.

Table II. Estimated intrinsic damage thresholds for Nd:doped laser glasses[a]

Glass	Threshold Energy (mJ)	Threshold Energy Density (KJ/cm^2)
A	20 ± 2	1.3 ± 0.3
B	18 ± 2	1.2 ± 0.3
C	18 ± 2	1.2 ± 0.3
D	20 ± 2	1.3 ± 0.3
E	20 ± 2	1.3 ± 0.3

[a]Data obtained by focusing radiation from a Q-switched Nd:glass laser (τ = 25 ns) with a 181-mm focal length lens. Estimated spot size radius of the 1/e point of intensity is 21 μm.

8.

2.4 Damage in LiNbO$_3$, Calcite, KDP, and KD*P [6]

An attempt was made to determine the mechanisms for self-focusing in LiNbO$_3$, calcite, KDP, and KD*P. LiNbO$_3$, KDP, and KD*P are electro-optic crystals used in the fabrication of Pockels cells. Calcite is a common polarizer material. The conclusion we reached was that inclusions are the principal cause of damage in these crystals, although filamentary damage caused by self-focusing was observed in LiNbO$_3$ at power levels significantly above the damage threshold. Energy densities in the vicinity of damage sites in these crystals were tabulated.

2.5 Damage in Neodymium-Doped Thoria:Yttrium Oxide Ceramic Laser Rod [6]

In cooperation with Charles Greskovitch of the General Electric Company Corporate Research and Development Center, we conducted bulk and surface damage threshold measurements on a laser rod constructed of neodymium-doped thorium:yttrium oxide ceramic. The damage thresholds relative to several optical materials were tabulated. The thresholds of this laser rod were lower than the thresholds of all the other materials tested with the exception of SF 55 glass.

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A. Feldman, D. Horowitz, and R. M. Waxler
"Relative Importance of Electrostriction and the Kerr
Effect to Self-Focusing in Optical Glasses"
Appl. Phys. Lett. 21, 260 (1972).

A. Feldman, D. Horowitz, and R. M. Waxler
"Relative Contribution of Kerr Effect and Electrostriction
to Self-Focusing" in Laser Induced Damage in Optical Materials:1972
NBS Special Publication 372
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A. Feldman, D. Horowitz, and R. M. Waxler
"Self-Focusing in Yttrium Aluminum Garnet and Optical Glasses" in
Laser Induced Damage in Optical Materials:1973
NBS Special Publication 387
(U.S. Government Printing Office, Washington, D.C., 1973) pp. 26-35.

A. Feldman, D. Horowitz, and R. M. Waxler
"Mechanisms for Self-Focusing in Optical Glasses"
IEEE J. Quantum Electron. QE-9, 1054 (1973).

A. Feldman
"Relations Between Electrostriction and the Stress-Optic Effect"
To be published.

A. Feldman, D. Horowitz and R. M. Waxler
"Stress-Optic Measurements in the Infrared"
To be published in Proceedings of the Conference on High-Power
Infrared Window Materials, 1973.

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[6] A. Feldman, D. Horowitz, and R. M. Waxler, "Laser Damage in Materials," National Bureau of Standards Report NBSIR 73-268 (August 1973).

[7] A. Feldman, D. Horowitz, and R. M. Waxler, "Self-Focusing in Yttrium Aluminum Garnet and Optical Glasses," in Laser Induced Damage in Optical Materials: 1973, NBS Special Publication 387 (U. S. Government Printing Office, Washington, D. C., 1973), pp. 26-35.

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[9] E. L. Dawes and J. H. Marburger, "Computer Studies in Self-Focusing," Phys. Rev. 179, 862 (1969).

[10] A. A. Maradudin and E. Burstein, "Relation Between Photo-elasticity, Electrostriction, and First Order Raman Effect in Crystals of the Diamond Structure," Phys. Rev. 164, 1081 (1967).

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16. ABSTRACT *(A 200-word or less factual summary of most significant information. If document includes a significant bibliography or literature survey, mention it here.)*

   This report summarizes the study of damage and self-focusing in materials used in Q-switch solid-state laser systems. In borosilicate crown glass, fused silica, dense flint glass, and yttrium aluminum garnet, self-focusing appears to be the main cause of damage. An analysis of damage threshold measurements with linearly polarized radiation and circularly polarized radiation suggests that the Kerr effect is the dominant self-focusing mechanism with a significant contribution to self-focusing from the thermal effect. The electrostrictive effect is negligible. The damage threshold in Nd:doped laser glasses appears to be intrinsic. In all the above materials, the damage threshold for circular polarization is greater than the damage threshold for linear polarization. In lithium niobate, calcite, potassium dihydrogen phosphate, and deuterated dihydrogen phosphate, damage at the lowest levels is caused by inclusions. Bulk and surface damage thresholds in nd:doped thoria:yttrium oxide ceramic are obtained relative to bulk damage thresholds in several optical materials. Relationships under different geometric boundary conditions are also derived for solid materials between the stress-optic coefficients and the electrostrictive coefficients.

17. KEY WORDS *(six to twelve entries; alphabetical order; capitalize only the first letter of the first key word unless a proper name; separated by semicolons)* Absorption coefficient; damage threshold; electrostriction; inclusion damage; intrinsic damage; Kerr effect; laser damage; laser glasses; laser rod materials; modulator crystals; nonlinear index of refraction; optical glasses; polarizer materials; self-focusing.

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