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Single- and multiple-pulse laser-induced bulk damage of fused and crystalline SiO_2 has been investigated. We studied the dependence of the intensity required to produce macroscopic damage upon wavelength, focal spot size, pulse duration, and pulse repetition frequency. It is found that the material must undergo some undetected microscopic change prior to macroscopic damage which increases the probability of damage with increasing number of pulses. The multiple pulse damage depends strongly on spot size, occurring in far fewer pulses at larger spot sizes than at small ones, and displays a significant dependence on pulse length and repetition frequency. Possible mechanisms are discussed.

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INTRODUCTION

The macroscopic damage of solids under repeated pulsed laser irradiation at intensities too low to cause damage in a single pulse involves mechanisms not evident in single-pulse laser-induced damage. Recent papers by the present authors[1] and others[2–5] have focused renewed attention on the accumulation of microscopic laser-induced changes in such materials prior to macroscopic damage. Such accumulation effects call into question the simple view of a sample being undamaged unless a visible region of melted or cracked material is produced. In alkali halides multiple irradiation at 10.6 μm revealed no visible microscopic damage, but calorimetric measurements showed that the absorption coefficient of the material increased irreversibly during multiple-pulse irradiation just prior to the onset of catastrophic damage.[2] As reported in Ref. 1, no changes in absorption or scattering of the irradiating laser beams were observed prior to macroscopic damage of SiO_2 by repetitively-pulsed 1.064- or 0.532-μm light. However, the results reported in Ref. 1 showed clear evidence of accumulated laser-induced changes in the material and a dependence of damage on wavelength and material, fused silica requiring fewer pulses at a given fraction of the single-pulse damage threshold than crystalline quartz.

In the present study we have extended the wavelength range to include 0.355 μm and examined the multiple-pulse damage dependence on focal spot size, pulse duration and pulse repetition frequency. Damage is defined by the appearance of a flash of light, the attenuation of the transmitted pulse and the production of a macroscopic zone of melted and cracked material, as observed by the scatter of a coaxial He–Ne laser beam. It is found that the single-pulse damage threshold decreases with decreasing wavelength, while the multiple-pulse damage behavior varies more strongly with material, spot size, pulse duration, and wavelength than does the single-pulse threshold. In addition, more pulses are needed to produce macroscopic damage at a given intensity under irradiation at a 1-Hz pulse repetition rate than at 10 Hz.

EXPERIMENT

The experimental apparatus used for damage studies has been described.[1] The light source was a Molectron Nd : YAG Q-switched laser providing 10 pulses per second of nearly Gaussian temporal and spatial shape, with frequency doubling and tripling crystals. The beam was tightly focused into the sample to generate bulk rather than surface damage. The incident and scattered energies of each pulse were monitored together with the incident and transmitted waveforms of the pulse producing catastrophic damage. Laser calorimetry was used to detect changes in the absorption coefficient of the sample prior to catastrophic damage, and to perform linear absorption measurements at several wavelengths. Nonlinear absorption measurements were also performed at 0.355 μm using the frequency tripled output of the Q-switched Nd : YAG laser.

The materials used in this work were Suprasil-1 and UV grade Corning 7940 fused silica and optical grade Sawyer single-crystal quartz. Samples of the latter were oriented such that the laser beam propagated along the z-axis in the 1.064-μm studies, but to avoid surface damage the samples were turned to provide a longer path length for the 0.532- and 0.355-μm measurements.

RESULTS AND DISCUSSION

To study multiple-pulse laser-induced damage quantitatively, it is first necessary to determine the threshold intensity required for single-pulse macroscopic damage. In this work, I_N is the average of the intensity above which damage always occurs in N or fewer pulses and the average of the intensity below which damage never occurs in N pulses. The width of the intensity region in which damage sometimes occured in N pulses is typically a few percent of the threshold intensity.

Table I summarizes the present results for single-pulse thresholds and is consistent with published focal volume and pulse duration dependences although the thresholds reported here are somewhat lower.[6–8] A rather strong wavelength dependence is evident in the results listed in Table I, particularly in that the 0.355-μm thresholds are significantly lower than the 0.532-μm results. The thresholds at 1.064 μm are generally comparable to those at 0.532 μm, although results for Corning 7940 may indicate a continued increase of damage threshold with wavelength. This trend is generally regarded to be indicative of multiphoton processes.[9] Cata-

TABLE I. Single-pulse damage thresholds. The listed parameters are the focal spot size at e^{-2} intensity, w, and the pulse duration, full width at half-maximum, t.

Material	$I_{\text{threshold}}$ (GW/cm^2) $\lambda = 1.064\,\mu$m	$I_{\text{threshold}}$ (GW/cm^2) $\lambda = 0.532\,\mu$m	$I_{\text{threshold}}$ (GW/cm^2) $\lambda = 0.355\,\mu$m
Crystalline Quartz (Sawyer optical grade)	130 ± 20 $w = 14.5\,\mu$m $t = 23$ ns	100 ± 15 $w = 7.0\,\mu$m $t = 14$ ns	45 ± 5 $w = 5.9\,\mu$m $t = 13$ ns
Suprasil-1 fused silica	110 ± 15 $w = 13.8\,\mu$m $t = 24$ ns	90 ± 15 $w = 6.6\,\mu$m $t = 15$ ns	35 ± 5 $w = 6.1\,\mu$m $t = 13$ ns
Corning 7940 fused silica	110 ± 15 $w = 13.8\,\mu$m $t = 24$ ns	110 ± 15 $w = 6.6\,\mu$m $t = 16$ ns	40 ± 5 $w = 5.0\,\mu$m $t = 14$ ns
	110 ± 15 $w = 15.6\,\mu$m $t = 19$ ns		
	120 ± 20 $w = 9.0\,\mu$m $t = 27$ ns		
	220 ± 40 $w = 7.2\,\mu$m $t = 18$ ns		

strophic self-focusing does not appear to control the present damage data.[10]

Figure 1 shows the results of low intensity (cw) absorption measurements on Corning 7940 and Sawyer optical quartz. Since surface absorption has not been separated from bulk absorption, the indicated absorption coefficients should be regarded as upper limits. The increased absorption in the ultraviolet may be a clue to the decrease in the damage threshold at 0.355 μm. No measurable nonlinear absorption was found at this wavelength. The larger absorption of crystalline quartz is most likely a surface quality effect.

Multiple-pulse-induced damage measurements were made on Corning 7940 at 1.064 μm under the same four sets of spot size, w, and pulse duration, t, as those shown in Table I. As shown in Fig. 2, by plotting the number of pulses N, to

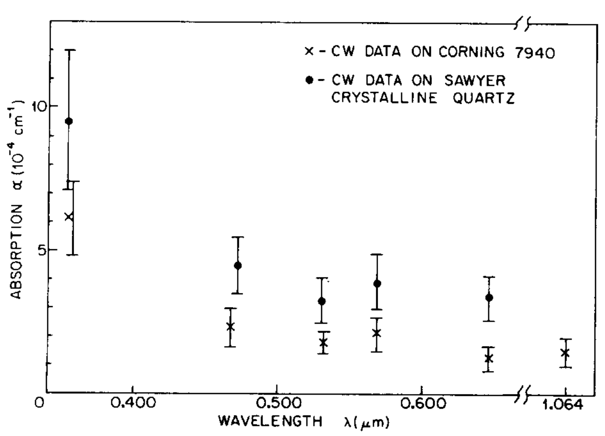

FIG. 1. Linear absorption coefficients of Corning 7940 fused silica, × and of Sawyer optical grade crystalline quartz, ●. The data have not been corrected for surface absorption.

which a site was exposed, against the intensity normalized by the single-pulse threshold intensity, one can determine whether multiple-pulse damage follows the same spot size and pulse length dependence as single-pulse damage. Note that the units of the abscissa in Fig. 2 are independent of spot size and pulse duration. For the short pulse, small spot case the data exhibit a rather clear threshold-like behavior. Indeed only one multiple-pulse damage event was observed at an intensity below the single-pulse threshold uncertainty, indicated by the dashed lines. The long-pulse, large spot-size data showed the behavior displayed in Ref. 1, where damage events occurred in increasing number of pulses at smaller subthreshold values. The shaded area enclosing all multiple-pulse damage events, indicates this trend. Table II summarizes the results by showing the fraction of threshold intensity at which N equals 1, 30, and 1000. It is evident from the table that a smaller fraction of threshold intensity is required to cause macroscopic damage in a given number of pulses when the pulse duration is longer and the spot size is larger. The spot size dependence is indicative of an initial concentration of damage promoting defects. The pulse duration dependence is consistent with the gradual accumulation of damage promoting microscopic changes in that fewer long pulses are needed to achieve damage than short pulses of the same intensity. The reason for plotting (Fig. 2) data characterized by the first ($w_1 = 7.2\ \mu$m $t_1 = 18$ ns) and last ($w_4 = 13.8\ \mu$m $t_2 = 24$ ns) sets of beam parameters, listed in Table II, in which both the beam diameter and the pulse length were changed, is for the purpose of demonstration of the damage behavior. When only the pulse length ($w_2 = 9.0\,\mu$m, $t_2 = 27$ ns) or only the beam diameter ($w_3 = 15.6\,\mu$m, $t_3 = 19$ ns) was changed smaller alterations were seen in the damage trend.

Multiple-pulse laser-induced damage data have also

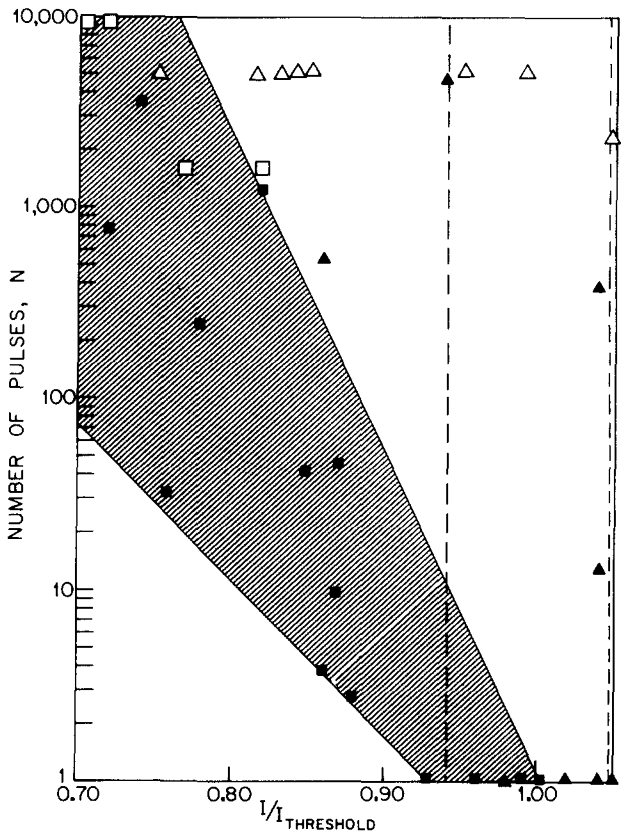

FIG. 2. Multiple pulse damage data on two samples of fused silica at 1.064 μm. Full symbols indicate an experiment where damage occurred in the indicated number of pulses, while open symbols indicate no damage. The squares represent data for sample A where the laser beams characteristics were $w = 13.8\,\mu$m and t (FWHM) = 24 ns. The triangles represent data for sample B where the laser beams characteristics were $w = 7.2\,\mu$m and t (FWHM) = 18 ns. The dashed line as well as the shaded area are explained in the text.

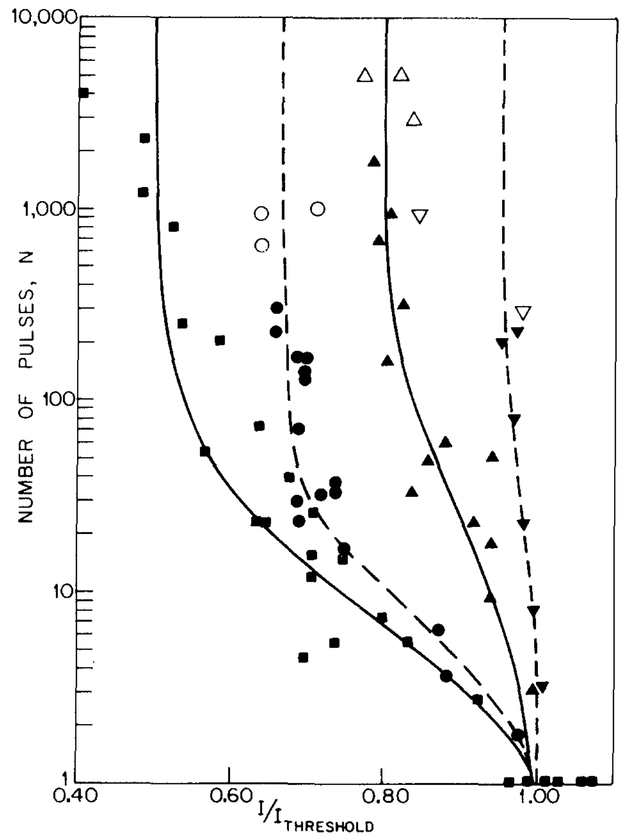

FIG. 3. Multiple pulse damage experiment on fused and crystalline SiO_2 at 0.355 μm. ■ : fused SiO_2, sample damaged, laser pulse repetition frequency = 10 Hz. ● : fused SiO_2, sample damaged, laser pulse repetition frequency = 1 Hz. ○ : fused SiO_2, sample did not damage, laser pulse repetition frequency = 1 Hz. ▲ : crystalline SiO_2, sample damaged, laser pulse repetition frequency = 10 Hz. △ : crystalline SiO_2, sample did not damage, laser pulse repetition frequency : 10 Hz. ▼ : crystalline SiO_2, sample damaged, laser pulse repetition frequency : 1 Hz. ▽ : crystalline SiO_2, sample did not damage, laser pulse repetition frequency : 1 Hz. The beam waist and the pulse duration (FWHM) were 5.0 μm and 14 ns for the fused SiO_2 and 5.9 μm and 13 ns for the crystalline SiO_2. See text for meaning of curve.

been taken on Suprasil-1 and Sawyer optical grade single-crystal quartz samples at 1.064 μm, on Sawyer quartz and both types of fused silica at 0.532 μm and on Sawyer quartz and Corning 7940 at 0.355 μm. The results are summarized in Table III. Comparison among the materials shows that one must stay at intensities considerably closer to threshold to achieve macroscopic damage within a given number of pulses in the crystalline quartz than in the Corning fused silica and Suprasil fused silica. This remains true at all wavelengths. These differences in multiple-pulse damage behavior contrast with the single-pulse damage thresholds which

TABLE II. Fraction of threshold intensity needed to cause multiple-pulse damage in Corning 7940 after 1, 30, and 1000 pulses at 1.064 μm.

w(μm)	t (ns)	$I_1/I_{\text{threshold}}$	$I_{30}/I_{\text{threshold}}$	$I_{1000}/I_{\text{threshold}}$
7.2	18	1.00	1.00	0.96
9.0	27	1.00	0.98	0.95
15.6	19	1.00	0.96	0.88
13.8	24	1.00	0.82	0.77

are similar for all the materials. This contrast suggests that different mechanisms control single- and multiple-damage.

A strong wavelength dependence is also evident in Table III. For all three materials multiple-pulse damage after a given number of pulses can be achieved at a smaller fraction of threshold intensity at 0.532 and 0.355 μm than at 1.064 μm. This trend is suggestive of color center generation or interaction of the light with submicron defects.

When macroscopic damage occurs after exposure to many virtually identical pulses the distribution of damage events implies that some minimum intensity or fluence is required to initiate the accumulation of microscopic damages or other changes which eventually lead to macroscopic damage. This was proven to be true in[4] for irradiation with the 1.064- and 0.532-μm light. Figure 3 shows the results for 0.355 μm for fused silica and crystalline quartz. At any given intensity significantly below threshold, there are no damage events in the first few pulses, then damage begins to occur frequently on later pulses. It thus appears that the sample is being changed by the repeated laser irradiation. To check the nature of these changes, i.e., whether they were permanent

TABLE III. Fraction of threshold intensity needed to cause multiple-pulse damage after 1, 30, and 1000 pulses for various samples and wavelengths. Pulse repetition frequency is 10 Hz.

Material and wavelength	$I_1/I_{\text{threshold}}$	$I_{30}/I_{\text{threshold}}$	$I_{1000}/I_{\text{threshold}}$
Corning, 1.064 μm ($w = 13.8\ \mu$m, $t = 24$ ns)[a]	1.00	0.82	0.77
Suprasil, 1.064 μm ($w = 13.8\ \mu$m, $t = 24$ ns)	1.00	0.89	0.86
Sawyer, 1.064 μm ($w = 14.5\ \mu$m, $t = 23$ ns)	1.00	0.96	0.92
Corning, 0.532 μm ($w = 6.6\ \mu$m, $t = 16$ ns)	1.00	0.63	0.58
Suprasil, 0.532 μm ($w = 6.6\ \mu$m, $t = 15$ ns)	1.00	0.68	0.64
Sawyer, 0.532 μm ($w = 7.0\ \mu$m, $t = 14$ ns)	1.00	0.86	0.70
Corning, 0.355 μm ($w = 5.0\ \mu$m, $t = 14$ ns)	1.00	0.66	0.56
Sawyer, 0.355 μm ($w = 5.9\ \mu$m, $t = 13$ ns)	1.00	0.94	0.82

[a] Chosen from Table II for comparison with the other two materials.

or not, the repetition frequency of the experiment was changed from 10 to 1 Hz. This was accomplished by placing a mechanical shutter in the beam path and allowing every 10th pulse from the 10-Hz laser to reach the sample. The results are shown in Fig. 3. Clearly, both Corning 7940 and Sawyer optical grade quartz require more pulses of a given peak intensity to damage at 1 Hz than at 10 Hz. Similar experiments performed on Corning 7940 at 0.532 μm indicate the same trend. The curves are eye guiding traces, solid for the 10-Hz data and dashed for the 1-Hz data. This observed dependence of multiple-pulse damage behavior on pulse repetition frequency confirms the conclusion that accumulated laser-induced changes in the sample lead to catastrophic damage. The sensitivity of the damage mechanism to such low frequencies is indeed surprising and it is being studied further. The dependence of the pulse repetition frequency effect on temperature is very probable and such studies are in preparation.

Multiple-laser-induced damage in SiO_2 observed in this work clearly indicates that macroscopic damage results from undetected microscopic changes induced by earlier pulses. A significant dependence of the damage threshold with wavelength, pulse duration, spot size, and repetition rate was observed. The dependence on spot size and repetition rate show that the defect responsible for laser damage has a distribution with a characteristic spacing of a few microns and has a lifetime of the order of a tenth of a second. These properties are suggestive of the role of such transient defects as color centers.

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