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Picosecond Damage in Y_2O_3 Stabilized Cubic Zirconia

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The hardness, strength, and transparency of cubic zirconia ZrO_2 makes it a good candidate as a laser window material; however, to obtain these qualities a relatively large percentage of Y_2O_3 is added to stabilize the crystals and increase their mechanical strength. As impurities are often regarded as the initiators of laser-induced damage we have performed a study of the damage thresholds of optical quality ZrO_2 stabilized with Y_2O_3. These thresholds have been measured with picosecond 1.06 µm laser pulses in crystals having Y_2O_3 concentrations of 9.4%, 12%, 15%, 18%, and 21%. We found that the addition of Y_2O_3 for increasing the mechanical strength of zirconia does not necessarily lead to a decrease in the damaging irradiance. In addition, the thresholds for ZrO_2 are comparable to those of NaCl.

Key Words: laser damage; picosecond pulses; ZrO_2 stabilized with Y_2O_3; estimation of n_2; 1.06 µm.

1. Introduction

The ideal optical materials for use with high power lasers should be transparent over a broad spectral range, mechanically strong, environmentally stable, and resistant to laser induced damage. Few materials meet all these criteria. For example, NaCl has low absorption from the infrared to the ultraviolet, is resistant to pulsed laser damage but has poor mechanical strength and is hydroscopic. ZnSe has good mechanical properties and broad band transmission but damages easily by pulsed laser irradiation. Fused silica is resistant to pulsed laser damage and has good mechanical properties but has high transparency only in the visible and near infrared spectral region.

Cubic zirconia stabilized with Y_2O_3 is mechanically strong, transparent from the UV to the mid-infrared and is now available in the industrial market. These factors make this material a good candidate for use as a high power laser window. The laser damage thresholds of this material had not previously been measured. In this paper we present measurements of the bulk laser-induced damage threshold of cubic zirconia stabilized with Y_2O_3 having concentrations of 9.4%, 12%, 15%, 18%, and 21%. Measurements were conducted using picosecond pulses at 1.06 µm.

2. Experiment

The laser source used in this study was a passively mode-locked, microprocessor-controlled, neodymium:yttrium aluminum garnet (Nd:YAG) oscillator-amplifier laser system operating at 1.06 µm. A single pulse of measured Gaussian spatial and temporal irradiance distribution was switched from the mode-locked train and amplified. The temporal pulsewidth was 45 psec [full width at half maximum (FWHM)]. The width of each pulse was monitored by measuring the ratio of the square of the energy in the fundamental (1.06 µm) to the energy in the second harmonic, produced in a $LiIO_3$ crystal. The ratio was calibrated by measuring the pulsewidth using an autocorrelation scan. Essential details are described in reference 1.

The laser half-angle beam divergence was 0.18 mrad. The beam divergence and the spatial beam spot size were determined by pinhole scans at several different positions along the propagation direction. A 37 mm focal length lens designed for minimum spherical aberration was used to focus the light into the bulk of the materials. The calculated focal spot radius using this lens is 7.2 µm (HW 1/e M in irradiance). A rotating half-wave plate, polarizer combination was used to vary the irradiance on the sample. The energy incident on the sample was continuously monitored by a sensitive photodiode peak-and-hold detector, and was calibrated with respect to a pyroelectric energy monitor. The incident beam polarization on the sample was changed from linear to circular by adding a quarter wave plate in front of the focusing lens. Each site was irradiated only once, and damage was defined by the observation of scattered light from a coaxial HeNe laser as viewed through a 10x microscope.

3. Experimental Results and Discussion

Tables 1 and 2 summarize the results of measurements of the laser induced breakdown threshold for ZrO_2 stabilized with Y_2O_3. The ZrO_2 samples having Y_2O_3 concentrations of 9.4%, 12%, 15%, 18%, and 21% were studied for linearly and circularly polarized light at 1.06 μm. The uncertainties listed in the tables of data are the relative errors obtained by the method used in reference 9. For all the data presented in tables 1 and 2 we used 45 picosecond (FWHM) pulses and a calculated 7.2 μm focused spot size (HW 1/e M in irradiance). The values given for the electric field are rms fields corresponding to the peak-on-axis irradiance. As indicated in the tables of data, increasing the percentage of Y_2O_3 does not significantly change the breakdown threshold. The breakdown fields for linearly and circularly polarized light are displayed in figure 2. As clearly represented by bar graphs, the breakdown field remains unchanged (to within ±10%) for Y_2O_3 concentrations ranging from 9.4 to 21%.

In this work, we carried out the same measurements for circularly as well as linearly polarized light in order to determine the effect of self-focusing on laser-induced damaged threshold measurements. For the data listed here, we have done the polarization dependence test (2,6) which insures the absence of self-focusing in bulk damage measurements. Figure 3 is the experimentally determined ratio of breakdown electric field for circular polarized light to that for linearly polarized light. The ratios are constant (within uncertainties of experiment), and approximately equal to 1 for all different concentrations.* This implies that for our experimental conditions self-focusing did not dominate the damage process in these measurements. Note that we carried out the same experiment for a different focused spot size (10 μm). The ratios increased by a factor of 1.2 to 1.5 for all the samples tested. The polarization dependence of self-focusing has already been seen experimentally in a variety of materials. For example, for NaCl the ratio of the critical powers for circular to linear polarization is 1.37 to 1.46 [6]. For various laser glasses this ratio is equal to 1.50 [10]. Therefore, we concluded that for the larger focal spot size self-focusing is controlling the damage process.

The critical power for self-focusing of a focused Gaussian beam within a Rayleigh range, is obtained from numerical solutions of the nonlinear wave equation [4], and is given by

$$P_2 = 3.77c\lambda^2/32\pi^2 n_2$$

where n_2 is the nonlinear refractive index in esu, λ is the laser wavelength, and c is the speed of light in vacuum. If for the 10 μm focused spot size, self-focusing is the dominant process (as seems to be indicated by the polarization dependence), then the measured breakdown power (P_B) should be the same as the critical power P_2. We used the values of the measured breakdown power and the equation above to calculate an estimated value of n_2 for the ZrO_2 samples at 1.06 μm. Using the assumption that $P_B \sim P_2$ we find that $n_2 = 8 \times 10^{-13}$ esu. This value is in agreement with the rough theoretical estimation of n_2 in the next paragraph.

Self-focusing is an induced lensing effect in materials resulting from the changes of the optical dielectric constant when an electric field is applied. In this case, the total dielectric constant can be written as

$$\varepsilon = \varepsilon_0 + \varepsilon_2 \langle E \cdot E \rangle$$

where $\langle E \cdot E \rangle$ is the time average of the square of the optical field. The induced polarization in the medium is

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*The 15% concentration sample was tested near the samples' edge for circularly polarized light where the optical figure of the front surface was poor. This probably accounts for the slightly greater than unity ratio for this sample shown in figure 3.

$$P = \chi^{(1)}E + \chi^{(3)} \langle E \cdot E\rangle E$$

where

$$\varepsilon_o = 1+4\pi\chi^{(1)} \quad \text{and} \quad \varepsilon_2 = 4\pi\chi^{(3)}$$

and the refractive index n is

$$n = \sqrt{\varepsilon} \sim n_o + n_2 \langle E \cdot E\rangle \quad .$$

In solid materials, the n_2 can be roughly estimated by using the argument given in reference 8. There it is assumed that the nonlinear polarization, $\chi^{(3)} \langle E \cdot E\rangle E$, becomes strong when the electrostatic energy of the applied optical field $\varepsilon_o/8\pi \langle E \cdot E\rangle V$ (V is the volume of the atom) is comparable to the electronic transition energy, $\hbar\omega_o$. The nonlinear polarization will then be comparable to the linear polarization so that

$$\frac{\chi^{(3)} \langle E \cdot E\rangle E}{\chi^{(1)} E} \sim \frac{\varepsilon_o/8\pi \langle E \cdot E\rangle V}{\hbar\omega_o} \quad .$$

With $n_2 = 2\pi\chi^{(3)}/n_o$, this gives

$$n_2 \sim \frac{n_o (n_o^2-1)}{16\pi\hbar\omega_o N}$$

where n_o is the linear index of refraction and N=1/V is the number density of atoms. For the ZrO_2 sample with n_o = 2.12 [11] and $\omega_o \sim 55 \times 10^{14}$ Hz, we estimate n_2 to be 10×10^{-13} esu which agrees with our experimental estimate of 8×10^{-13} esu. The validity of this estimation procedure can be shown by comparing its predictions for other materials where n_2 has been measured. The estimate of n_2 for NaCl is 1.3×10^{-13} esu and for SiO_2 it is 1.0×10^{-13} esu which compares to experimentally determined n_2's of $(1.37 \pm .15) \times 10^{-13}$ esu and $(.62 \pm .03) \times 10^{-13}$ esu, respectively [7]. Another empirical expression which has accurately given the nonlinear refractive index for a wide variety of transparent insulating materials is

$$n_2\ (10^{-13}\ \text{esu}) = K \frac{(n_d-1)(n_d^2+2)^2}{\nu[1.517 + (n_d^2+2)(n_d+1)\ \nu/6n_d]^{1/2}}$$

where ν is the Abbe number, n_d is the helium d line refractive index and K=68 [12]. This expression predicts an n_2 for ZrO_2 of 13×10^{-13} esu which again agrees with our estimate.

4. Summary

Laser induced breakdown was studied at 1.06 μm for cubic zirconia stabilized with Y_2O_3. Samples having concentrations of 9.4%, 12%, 15%, 18%, and 21% were investigated. We find that increasing the percentage of Y_2O_3 to stabilize the zirconia does not decrease the breakdown threshold. The breakdown field is unchanged (to within ±10%) for Y_2O_3 concentration ranging from 9.4% to 21%. The breakdown fields for circular and linear polarized light were found to be approximately the same for the small spot size used. The implication of this is that for these conditions self-focusing did not play a major role in the measurements. Also, we estimate the nonlinear index of refraction n_2 for the ZrO_2 samples. For this estimate we used the breakdown data for which self-focusing was a dominant process. The value for n_2 agrees with theoretical estimates.

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5. References

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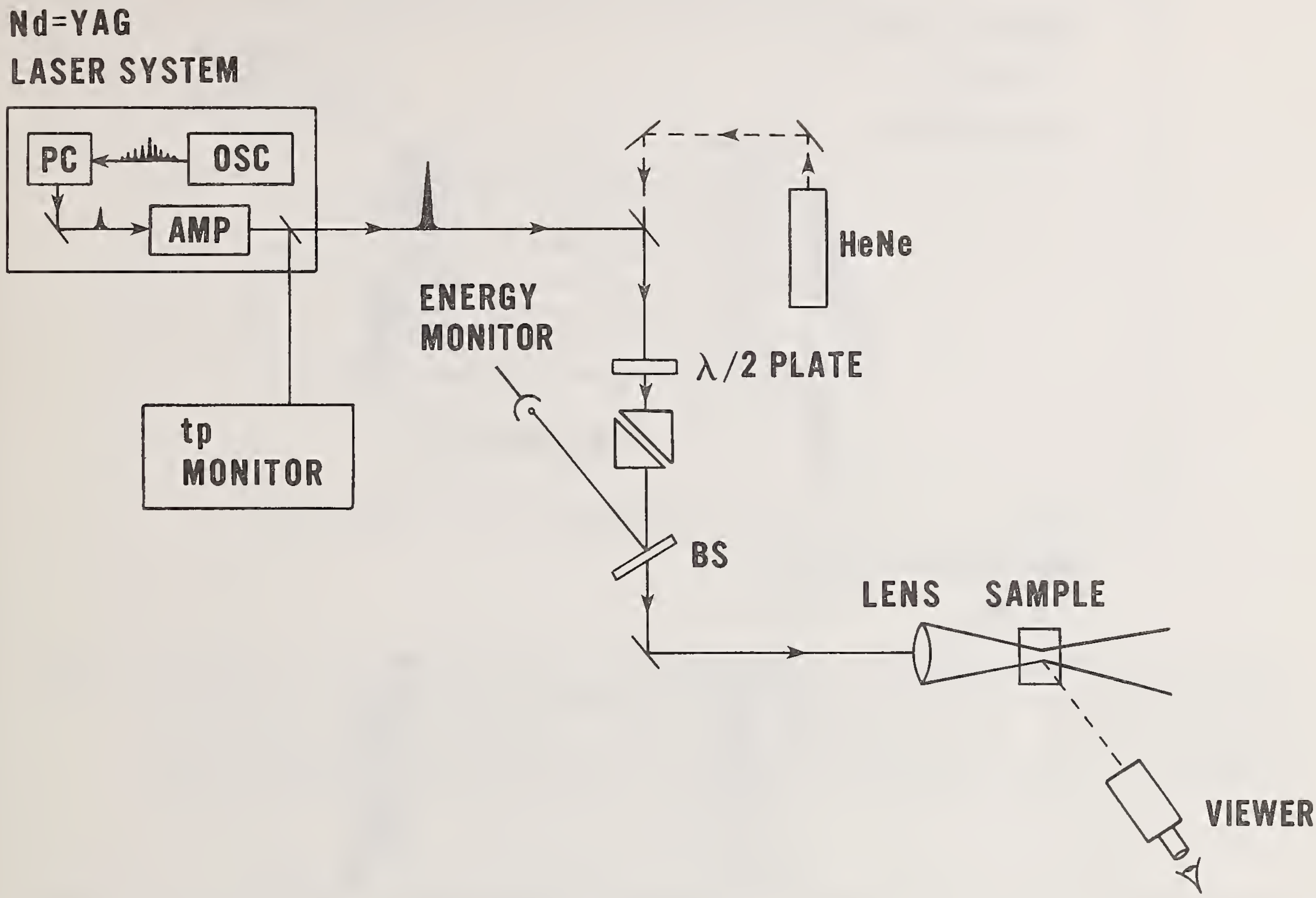

Figure 1. Experiment.

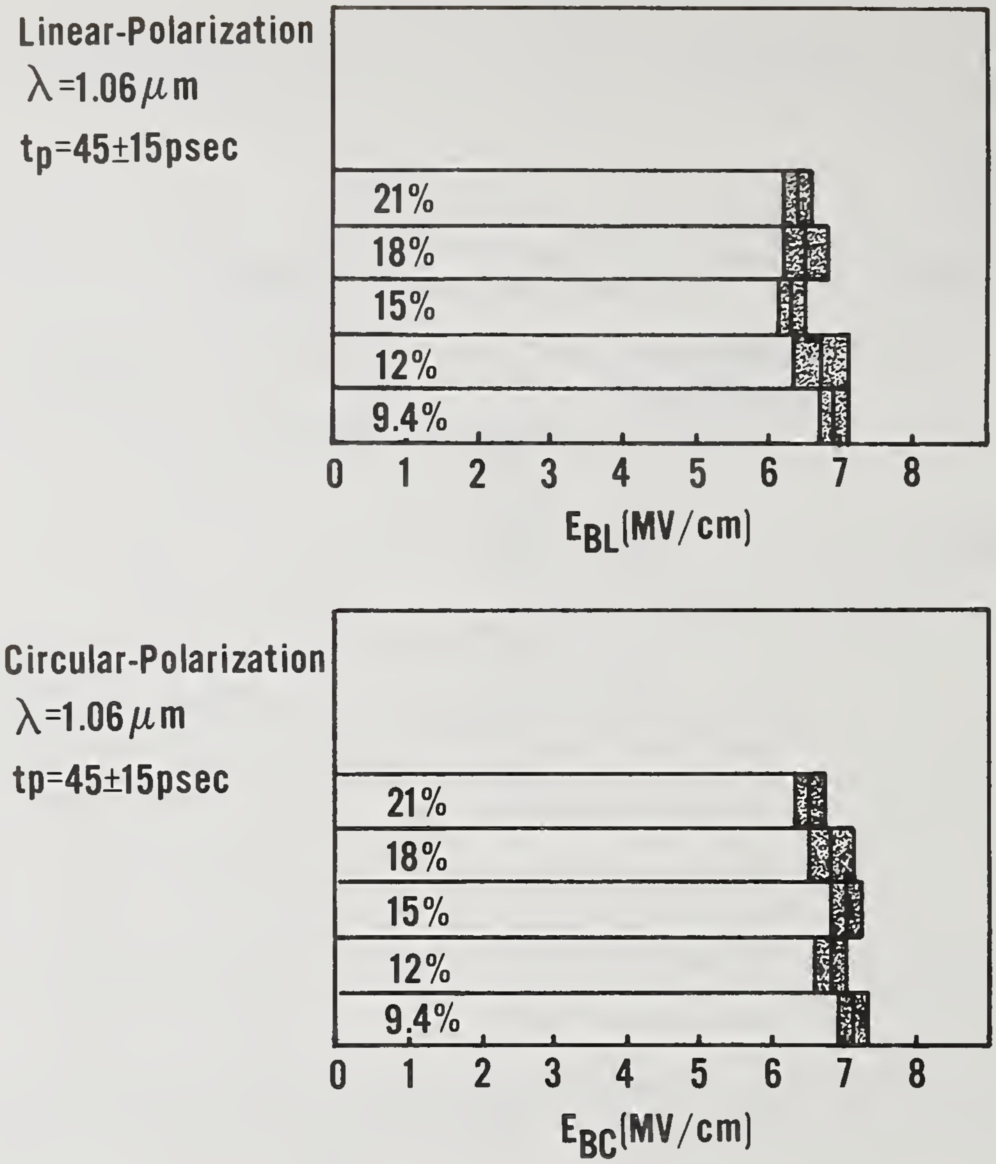

Figure 2. Experimentally determined breakdown electric field for linearly and circularly polarized light, E_{BL} and E_{BC}, for cubic zirconia ZrO_2 stabilized with Y_2O_3 having concentrations of 9.4%, 12%, 15%, 18%, and 21%.

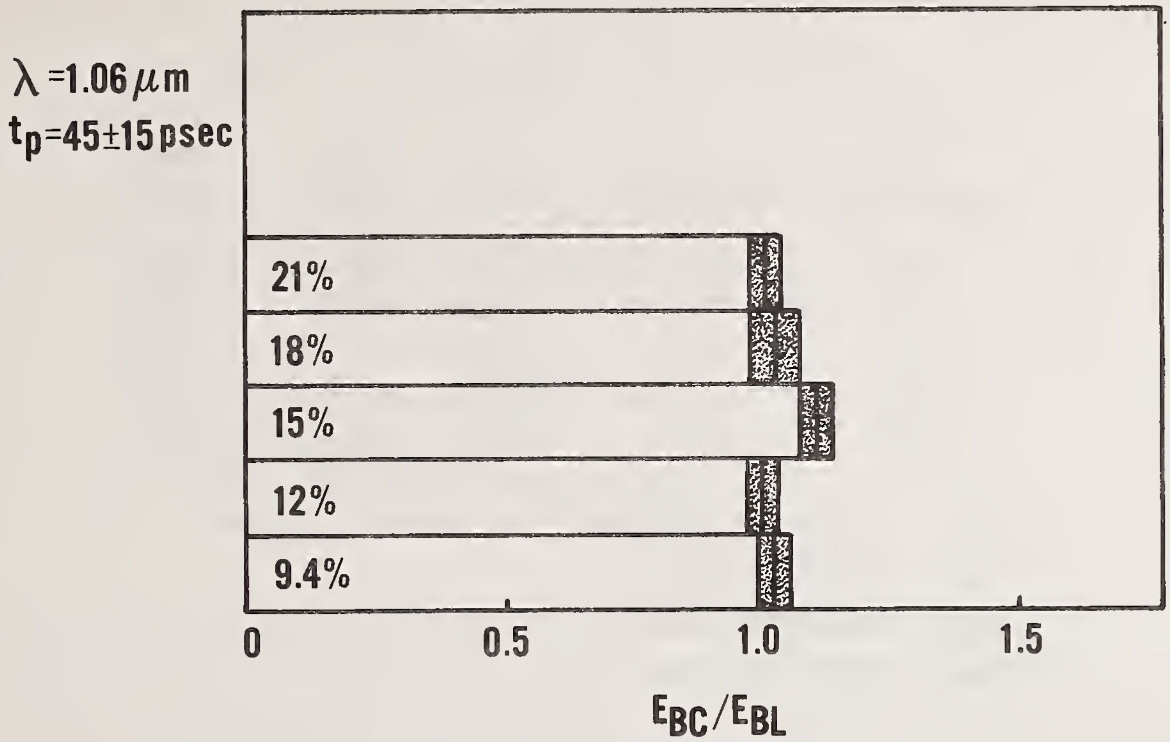

Figure 3. Experimentally determined ratio of breakdown electric field for linearly and circularly polarized light, E_{BC}/E_{BL}, for cubic zirconia stabilized with Y_2O_3 having concentrations of 9.4%, 12%, 15%, 18%, and 21%. (E_{BC} is the breakdown field for circularly polarized light and E_{BL} is the breakdown field for linear polarized light.) Note that $E_{BC} \sim E_{BL}$. The lack of polarization dependence indicates that its breakdown power is significantly less than its critical power for self-focusing.

Table 1. Laser induced damage data for ZrO_2 stabilized with Y_2O_3 at 1.06 μm using linearly polarized light.

Y_2O_3 Concentration	I_{BL} GW/cm^2	E_{BL} MV/cm	P_{BL} KW	F_{BL} J/cm^2
9.4%	265±18	6.9±0.2	438±30	12.7±0.9
12.0%	257±30	6.7±0.4	423±49	12.3±1.4
15.0%	222±11	6.3±0.2	366±19	10.6±0.6
18.0%	237±24	6.5±0.3	389±40	11.3±1.2
21.0%	230±17	6.4±0.2	380±28	11.0±0.8

I_{BL} = breakdown irradiance (peak on-axis irradiance)
E_{BL} = breakdown field
P_{BL} = breakdown power
F_{BL} = breakdown fluence

Table 2. Laser induced damage data for ZrO_2 stabilized with Y_2O_3 at 1.06 μm using circularly polarized light.

Y_2O_3 Concentration	I_{BC} GW/cm^2	E_{BC} MV/cm	P_{BC} KW	F_{BC} J/cm^2
9.4%	282±17	7.1±0.2	465±27	13.5±0.7
12.0%	260±17	6.8±0.2	430±28	12.4±0.8
15.0%	278±13	7.0±0.1	458±22	13.3±0.6
18.0%	257±25	6.8±0.3	424±42	12.3±1.2
21.0%	238±16	6.5±0.2	393±26	11.4±0.7

I_{BC} = breakdown irradiance (peak on axis irradiance)

E_{BC} = breakdown field

P_{BC} = breakdown power

F_{BC} = breakdown fluence