Statistical study of single and multiple pulse laser-induced damage in glasses

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Abstract: Single and multiple pulse laser damage studies are performed in Suprasil silica and BK-7 borosilicate glasses. Experiments are made in the bulk of materials at $1.064\mu m$ with nanosecond pulses, using an accurate and reliable measurement system. By means of a statistical study on laser damage probabilities, we demonstrate that the same nano-precursors could be involved in the multiple shot and single shot damage process. A damage mechanism with two stages is then proposed to explain the results. Firstly, a pre-damage process, corresponding to material changes at a microscopic level, leads the precursor to a state that can induce a one-pulse damage. And secondly a final damage occurs, with a mechanism identical to the single shot case. For each material, a law is found to predict the precursor life-time. We can then deduce the long term life of optical elements in high-power laser systems submitted to multipulse irradiation.

OCIS codes: (140.3330) Laser damage; (140.3440) Laser-induced breakdown; (160.2750) Glass and other amorphous materials; (160.6030) Silica

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1. Introduction

In some optical materials, laser-induced damage threshold can decrease with multiple pulse irradiation. This phenomenon, often called "fatigue" laser-induced damage, has been observed and studied in different transparent materials such as polymers [1], crystalline solids [2, 3, 4] and glasses [6, 7, 8, 9, 10, 11, 12]. It is a strong limitation for many high-power laser applications, where optical elements are submitted to a large number of pulses. A complete review of this subject has been made by Chmel [18], about experimental data and proposed multi-shot mechanisms. From this work, it comes out that despite considerable studies, multiple pulse laser damage in glasses remains incompletely understood. And at present, there is not a commonly accepted and demonstrated mechanism of the multi-pulse subthreshold laser damage.

To contribute in the understanding of this phenomenon, we propose a new approach of this subject. By means of a statistical study, we plot multipulse laser damage threshold curves that we analyze thanks to a phenomenological model. The use of this model added to accurate threshold curves plotting permits to obtain a reliable estimate of threshold as a function of the number of cumulated shot. These results will give us good arguments to emphasize possible mechanisms.

We chose to perform experiments on Suprasil silica glasses, known for their high damage threshold, and on BK-7 borosilcate glasses which are commonly used in laser applications.

Firstly, experimental apparatus and test procedure are described. Secondly, we present single and multiple pulse laser damage probabilities, obtained at 1.064μm with variation of cumulated shots up to 1000. To finish, results are discussed and compared with possible mechanisms for multiple pulse damage.

2. Materials and Methods

2.1 Experimental setup

The apparatus used for laser damage experiments has been described in details elsewhere [19], and only a brief description is given here. The set-up involves a single mode YAG laser beam with 1.064μm wavelength and 7ns pulse duration (figure 1). Variable attenuation is provided with a rotating half wave plate/polarizer combination. Energy of the incident beam is measured with a calorimeter and a pyroelectric-detector. Beam profile of the focused beam is analyzed with an optical system associated to a CCD camera. In this work the laser beam is focused with an adapted objective into the bulk of the sample with a spot size of 12μm (defined by the diameter at which the fluence is e^{-2} times its maximum value). The depth of focus in this configuration is about 100μm, which avoids any interface interaction. The sample is observed by an *in situ* optical microscope, which ensures a real time observation of the irradiated zone. "Damage" is defined as having occurred when a visible modification is detected with the microscope.

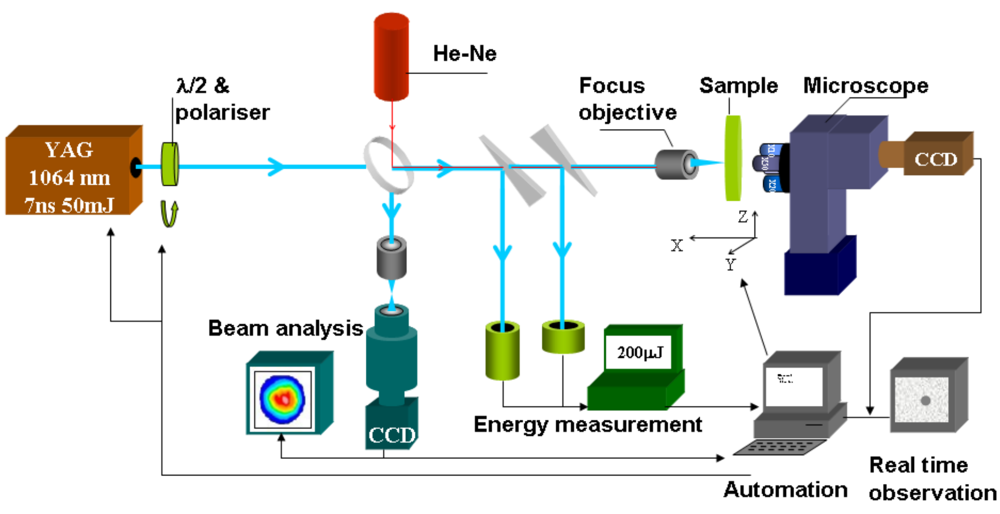

Fig. 1. Experimental set-up for laser damage testing

The whole apparatus is completely automatic. A computer controls sample displacement, the variable attenuation, laser shot synchronization and repetition rate. Energy and image of the focused beam are also acquired and recorded for each shot. In addition, an image processing has been implemented to obtain an accurate and real time detection of damage [19]. The simultaneous measurement of energy, beam profile, and

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damage detection by image analysis can be made for shot frequencies up to 20Hz.

2.2 Beam characterization

Multi-shot damage testing requires a highly stable test laser and the ability to check with accuracy each laser pulse. From this perspective, energy and image of the focused beam are associated and recorded for each shot. After each test sequence, these data are automatically analyzed. Parameters like beam diameter, maximum of energy, total energy, peak location, centroid are measured. Any malfunction is then detected and the corresponding shot can be rejected. This procedure permits to ensure constant laser performance throughout the test. Typical variations of the focused beam during 1000 shots are shown in table 1.

Table 1. Stability of the focused beam during 1000 shots at 10Hz. Beam waist is $12\mu m$ at 1064nm, with a pulse length of 7ns.

	Pointing	Beam waist	Energy
Maximum variation	$2.6\mu m$	6.9%	6.1%
Mean variation	$0.7\mu m$	1.7%	0.8%
Standard deviation	$0.4\mu m$	1.3%	0.7%

The accurate determination of the fluence on the sample is not immediate. Indeed, the beam shape does not have a perfect Gaussian profile and energy can be found outside the central peak. However by measuring the real beam profile, we can estimate the real fluence on the sample. This method, based on an image processing, has been described in ref. [19] and is systematically used in this work.

2.3 Test procedure

Single-shot damage measurements are made using the 1-on-1 procedure [20]. For each fluence, a single shot is delivered at thirty different areas on the sample, and a distance of 0.5 mm between each site is chosen to avoid interaction between tested zones [19]. The damage probability is given by the ratio $p = n/30$, where n is the number of damaged zones. In these conditions, the accuracy of the measurement is $\Delta p/p \approx 0.18$ [21].

For multi-shot damage measurements, two different test procedures can be used : S-on-1 [22] or R-on-1 [23]. The R-on-1 procedure is often used by the laser damage community for multi-shot damage testing but appears difficult to interpret. Indeed, two parameters are involved : the number of shots on each site and the energy step between each shot. Consequently, in this work we used exclusively the S-on-1 procedure to measure the influence of the number of shots on the laser-induced damage threshold.

The experiment consists in exposing an unirradiated zone to a train of pulses, until damage is detected or until the number of pulses reached N without damage (N=10, 100, or 1000). When damage is detected, the laser is immediately stopped to avoid damage growth and subsequent pollution of the sample. Then, for each fluence the damage probability is given by the ratio $p = n'/30$, where n' is the number of damaged zones in less than N shots.

3. Experimental results for silica and borosilicate glasses

The samples studied in this work are a BK-7 borosilicate glass sample, chosen for its common use, and a Suprasil silica sample, which is the glass having the best threshold value at this wavelength [19].

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As we have explained previously, damage mechanisms in these two materials remain unclear. In order to simplify the study, we chose in first time to concentrate our efforts on the bulk of materials. Indeed, in this case we avoid any interface effects due to the polishing process, surface contamination...

3.1 Single-shot laser damage threshold curves

Threshold curves measured in the conditions described above are presented on figure 2 for the materials under study.

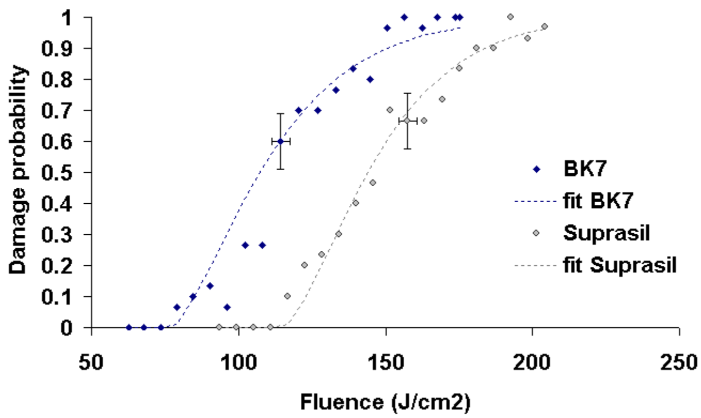

Fig. 2. Threshold curves measured in the bulk of BK7 and Suprasil, in 1-on-1 mode at 1064nm.

The shape of these curves is dependent on the spot size and on the material inhomogeneity : defects included in the bulk of material are assumed to be precursors of damage [8, 15, 16, 17]. A phenomenological model has been developed to analyzed these curves [24, 21], and can be described as follows. The damage probability is turned into the presence probability of a precursor receiving a fluence greater than its intrinsic threshold "T". In the case of a Gaussian illumination at normal incidence, a complete calculation (detailed in ref[21]) leads to the damage probability (P) as a function of the incident fluence (F), when F>T :

$$P(F) = 1 - exp(-dV_T(F)) \tag{1}$$

where d is the bulk density of precursors, and V_T is the efficient bulk where the energy density is greater than the precursor threshold T. V_T is defined as :

$$V_T = \frac{4}{6}\pi w_0^2 2z_R[u + \frac{u^3}{6} - \arctan(u)] \tag{2}$$

where w_0 is the beam waist, z_R the Rayleigh length and u :

$$u = \sqrt{\frac{F}{T} - 1} \tag{3}$$

Fits of the experimental curves by this model are shown on figure 2 (dotted lines). Low threshold values obtained are $115J/cm^2$ for Suprasil and $76J/cm^2$ for BK7. For both glasses a density of few tens of precursors in a $(100\mu m)^3$ volume is found.

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3.2 Multi-shot laser damage data

Multiple pulse damage measurements are made in the bulk of Suprasil and BK7 according to the procedure described in section *2.3*, with a repetition rate of 10Hz. Notice that no difference is observed between experiments at 1Hz or 10 Hz. Results are reported in table 2 for BK7 and table 3 for Suprasil : P_i corresponds to damage probability after i shots, with i =1, 10, 100 or 1000.

Table 2. Damage probability in the bulk of BK7 after i shots (P_i), with i=1,10, 100 or 1000. Beam waist is 12μm at 1064nm, with a pulse length of 7ns and a repetition rate of 10 Hz.

Fluence (J/cm^2)	P_1	P_{10}	P_{100}	P_{1000}
38	0	0	0	0
62	0	0.05	0.07	0.5
77	0.03	0.1	0.15	0.55
83	0.1	0.59	0.79	0.88
89	0.15	0.67	1	1
116	0.65	1	1	1

Table 3. Damage probability in the bulk of Suprasil after i shots (P_i), with i=1,10, 100 or 1000. Beam waist is 12μm at 1064nm, with a pulse length of 7ns and a repetition rate of 10 Hz.

Fluence (J/cm^2)	P_1	P_{10}	P_{100}	P_{1000}
65	0	0	0	0
77	0	0	0	0.03
85	0	0	0.03	0.3
95	0	0	0.17	0.7
105	0	0.03	0.4	0.9
113	0	0.13	0.8	1
122	0.17	0.3	0.9	1
133	0.2	0.53	1	1
144	0.43	0.77	1	1
162	0.47	0.9	1	1
182	0.57	0.97	1	1

The experimental data show that the damage threshold is strongly dependent on the number of shots, as it is well known in multiple pulse damage studies. The low threshold of BK7 and Suprasil for damage within 1000 shots is found to reduce about 30% to 40% compared with the single shot threshold. These results are consistent with other studies in similar materials [13, 14, 8].

Moreover, the damage morphology observed in the case of multiple shots is similar to the case of a single shot. In addition, the catastrophic breakdown occurs without any visible previous modification, as we can see on figure 3.

As for the single shot data, multiple pulse damage measurements reveal a dispersion between a low and a high threshold, which can be attributed to heterogeneities in the materials. The use of a small beam size in these experiments (12μm at $1/e^2$) has permits to highlight the statistical nature of the phenomenon. We discussed of possible mechanisms in the next section.

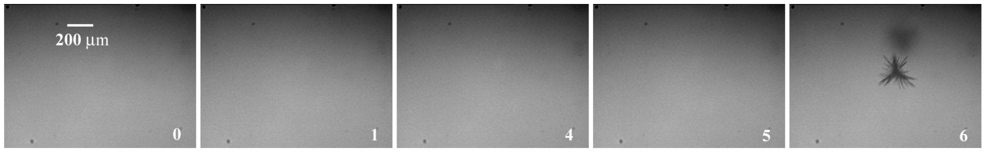

Fig. 3. Images after successive shots in the bulk of a Suprasil glass at $110J/cm^2$. Damage appears after the 6th shot, without any visible modification on the previous shots.

4. Discussion

Thanks to our specific set-up, we have obtained a large number of reliable data of multiple shot damage threshold, with a variation of the number of cumulated shots from 1 to 1000. Thus, it is possible to have a fresh look on multi-pulse damage in glasses. For instance, threshold dispersion can not be attributed to laser pulse-to-pulse fluctuations as in ref [8]. Real time detection of damage by image analysis permits to detect the very first damage created, and avoid any doubtful cases because of the use of less efficient diagnostics as in ref [1]. In addition the large number of data collected thanks to the automation avoid incorrect conclusions that can lead to discrepancies with subsequent measurements (as in refs [6, 7] and [10]).

4.1 Role of precursors centers in the repetitive shot damage process

In the case of single shot measurements, the threshold dispersion mentioned previously can be attributed to the presence of precursors which initiate damage in the material (described in section *3.1*). Then, the similar threshold dispersion observed in the case of multiple shot irradiation in glasses, can also be explained by the presence of defects or impurities which may play a role in the damage process. Two different hypothesis can be assumed : either these precursors are the same in the single and multiple shot cases or they are different.

In the case of the same precursors of damage for single and multiple shot in glasses, the existence of an accumulation effect during multi-pulse irradiation means that the intrinsic threshold of precursors called "T" in section *3.1* depends on the number of pulses "N" :

$$T = T(N) \tag{4}$$

Then the damage probability in the bulk of the material can be expressed as :

$$p(F) = 1 - e^{-d\frac{4}{3}\pi w_0^2 z_R \left[\frac{\left(\sqrt{\frac{F}{T(N)}-1}\right)^3}{6} + \left(\sqrt{\frac{F}{T(N)}-1}\right) - \arctan\left(\sqrt{\frac{F}{T(N)}-1}\right)\right]} \tag{5}$$

where d is the density of precursors. This density is known, since it has been determined previously with the fit of single shot threshold curves. Therefore we can verify if this simple model is adapted, by fitting experimental multiple shot threshold curves. In figure 4 for Suprasil and in figure 5 for BK7, we plot experimental damage probabilities as a function of fluence, for different cumulated shots "N" (N=1, 10, 100, 1000), as well as fits of these data with equation 5 (the free parameter is T(N)).

On these curves, we can observe a very good agreement between the measurements and the model described previously. This result permits to assume that our hypothesis of the same precursors for the single and multiple shot process is valid for Suprasil and BK7.

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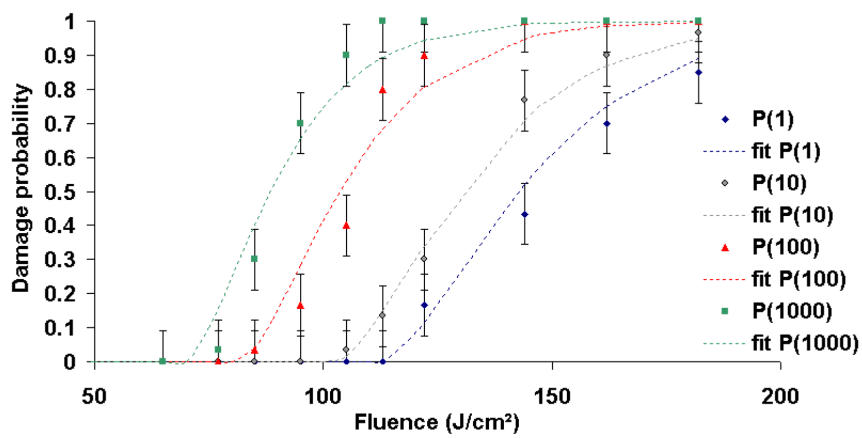

Fig. 4. Laser damage probability curves after 1, 10, 100, 1000 shots, respectively P(1), P(10) P(100), P(1000), measured in the bulk of Suprasil at 1064nm with a spot size of $12\mu m$. Data are fitted with the model presented in the text.

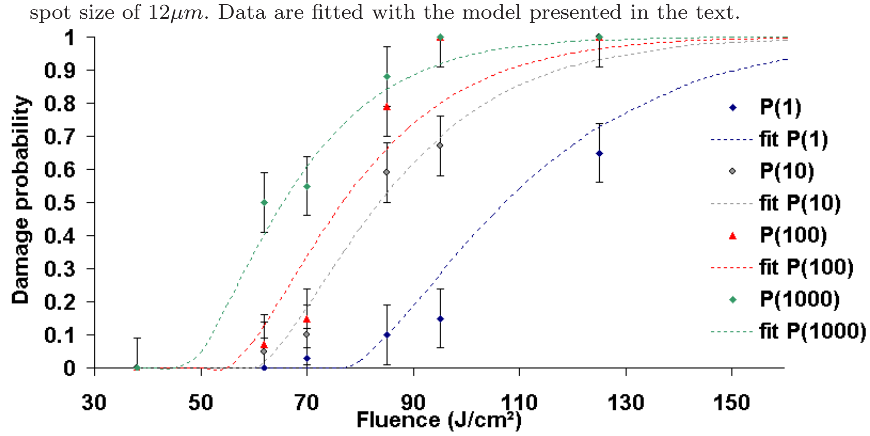

Fig. 5. Laser damage probability curves after 1, 10, 100, 1000 shots, respectively P(1), P(10) P(100), P(1000), measured in the bulk of BK7 at 1064nm with a spot size of $12\mu m$. Data are fitted with the model presented in the text.

4.2 Nano-precursor lifetime

Fits of the experimental data by the model permit to determine the intrinsic threshold of precursors T as a function of the number of shots N, giving an expression of precursor lifetime. These values are plotted on figure 6.

We can notice that these values can be fitted with a logarithmic law. This permits to obtain an empirical expression of precursor threshold as a function of the number of shots :

$$T(N) = -6,5 * ln(N) + 115 \, for \, Suprasil \quad (6)$$
$$T(N) = -4 * ln(N) + 76 \, for \, BK7 \quad (7)$$

These expressions of threshold as a function of N are of practical interest to predict the long term life of optical elements in high-power laser systems.

Notice that the test procedure applied to these two glasses, can be easily extend to other materials. Therefore we can give an adapted empirical law of precursor life time which depends of mechanisms. For instance, in the case of materials subjected to conditioning effects such as the bulk of KDP [27, 28, 29], a positive slope could be expected for the precursor lifetime as a function of N (figure 6).

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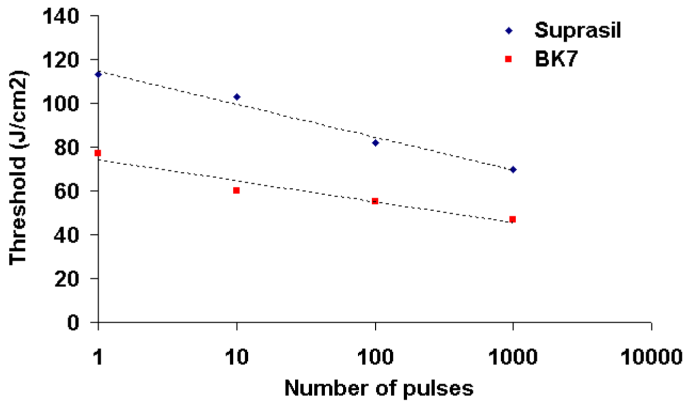

Fig. 6. Multi-shot low damage thresholds of Suprasil and BK7, obtained by fitting experimental data with equation 5. Dotted lines are logarithmic fits of these results.

However our study is made with a variation of N up to 10^3, for which the previous results are valid. The question asked is if these laws hold for millions of pulses. Different studies argued in favour of a "safe" (non-damaging) fluence at which no large number of pulses could produce macrodamage [10, 9], but we have to notice that this "safe" fluence has never been really demonstrated and measured given the limited number of pulses involved in all studies on this subject (up to 10^4). Indeed, measurements are very long for this kind of study, and a statistic on a large number of sites is necessary to obtain exploitable results. One solution could be found by using higher pulse repetition frequencies, but the time between each shot could have an effect on mechanisms.

Anyway, in the range of number of cumulated pulses "N" that we study, the logarithmic dependence of precursor lifetime as a function N that has been found, can be helpful to identify or reject potential mechanisms for the phenomenon of "fatigue" damage in glasses. We discuss this point in the next section.

4.3 Pre-damage mechanism

We showed that the precursors involved in the multiple shot damage process can be the same as the ones involved in the single shot process. We also observed that no precursory signs of damage can be detected before the catastrophic damage, which is similar to the single shot damage. A damage mechanism in two stages is proposed to explain the results. Firstly, a pre-damage process, corresponding to material changes at a microscopic level, leads the precursor to a state that can induce a one-pulse damage. And secondly a final damage process which is identical to the single shot case.

Different hypothesis are formulated for the pre-damage process, such as bond-breaking model[12], heating inclusion model[17], colored center model[4]... Nevertheless, none of these theories has been demonstrated to be responsible for the subthreshold damage mechanism in glasses. Difficulty in these studies are the lack of knowledge about these nano-centers, at the origin of the damage process. In order to resolve problems concerning the understanding of this phenomenon, a solution can be found in the study of "model" samples containing artificial nano precursors with known properties[26]. Irradiation with multiple shots in these samples and comparison of experimental data with numerical simulation will be of essential interest for the understanding of fatigue laser-induced damage in transparent materials.

5. Conclusion

We presented a study of multiple pulse laser damage in the bulk of glasses. Thanks to a specific apparatus and test procedure, we characterized accurately the laser damage threshold probabilities of Suprasil silica and BK7 borosilicate submitted to cumulated shots. All data were analyzed via a statistic approach, involving precursor densities, threshold, number of shots, and spot size, under the assumption that nano-precursors are responsible for laser damage. For these materials, we demonstrated that the same nano-precursors can be involved in the multiple shot and single shot damage process. The whole result let us assume a multipulse damage mechanism in two stages. It is composed of a pre-damage process, corresponding to material changes at a microscopic level, that leads the precursor to a state that can induce a one-pulse damage. And secondly a final damage process which is identical to the single shot case.

Thanks to the model, laws have been found to predict the precursors life-time and so the long term life of optical elements in Suprasil and BK7. To go further, different materials will be tested to highlight different behaviors and mechanisms, such as conditioning effects. The case of surfaces has a major interest for the community and will also be treated.

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