Assessment of mono-shot measurement as a fast and accurate determination of the laser-induced damage threshold in the sub-picosecond regime

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Standard test protocols need several laser shots to assess the laser-induced damage threshold of optics and, consequently, large areas are necessary. Taking into account the dominating intrinsic mechanisms of laser damage in the sub-picosecond regime, a simple, fast, and accurate method, based on correlating the fluence distribution with the damage morphology after only one shot in optics is therein presented. Several materials and components have been tested using this method and compared to the results obtained with the classical 1/1 method. Both lead to the same threshold value with an accuracy in the same order of magnitude. Therefore, this mono-shot testing could be a straightforward protocol to evaluate damage threshold in short pulse regime.

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Since the advent of the short pulse amplification technique in 1985 [1], short pulse high power laser facilities have been developed all around the world. The laser damage resistance of the optical components is of main concern as it always limits the overall system performances. Some ultra-intense high energy systems [2] use large apertures to transport this large amount of energy, thus reducing the energy density and then limiting laser damage of the optical surfaces. Most of the laboratories use small size optics for cost and space availability reasons. In both cases, it is necessary to increase the laser damage resistance of the optics. Current research is devoted to the development of new technologies to manufacture high reflective dielectric structures that exhibit high damage thresholds. The definition of new stack designs that reduce the enhancement of the electric field in the structure is one way to achieve an increase of the laser resistance [3,4]. Another working angle is the exploration of new dielectric materials, such as mixture, which shows high damage threshold and good optical properties (a high index of refraction for example) [5] and can be included in the multi-layer dielectric coating designs of reflective components.

To assess the laser-induced damage threshold (LIDT) of these materials or structures, ISO standards [6] are usually used. 1/1 procedure is a classical test to quantify the damage resistance of components at an irradiation, whereas S/1 procedure is devoted to the study of fatigue with multiple irradiations. Damage mechanism in the short pulse regime is known to be driven by nonlinear processes, such as multiphotonic ionization and impact ionization. It leads to a deterministic damage behavior in the case of a defect-free material for which its threshold can be linked to the bandgap, the pulse duration [7,8], and the distribution of the electric field in the stack in the case of optical interference coatings [9]. For both tests, standards recommend the test of at least 10 sites per fluence of irradiation. A step between each tested site is introduced to avoid cooperative effect (incubation [10] of the next site or ejection of particles) and to guarantee that the LIDT is established on pristine locations. This step is chosen to be four times the beam diameter as suggested by ISO standards. For a beam diameter around a hundred of micrometers, it represents a final consumed area in the order of a square centimeter. On the other hand, the rasterscan technique is useful to assess the quantity of defects of an optic that drastically reduce its laser resistance and, thus, its operating fluence [11]. In this Letter, we rather focus on the former protocols, i.e., the measurement of LIDT, intrinsic to materials. Since the optical components can be expensive or their manufacture a technical challenge, the problem is how to measure the LIDT of a component while consuming the minimum surface. We herein detail the principle of a mono-shot measurement in the short pulse regime using a commercially available laser: we report a simple and reproducible method for laser testing of an optical surface with only one shot. The LIDT is measured with a micrometer scale size beam by correlating the damage morphology with the local fluence in the laser beam. (A similar method using
a large beam has already been employed for the determination of damage densities [12]. This method has been tested on several dielectric coatings, and the results have been compared with the standard 1/1 procedure: a small difference between the two sets of results confirms the accuracy of the mono-shot method.

The experimental setup uses an S-pulse Amplitude Systèmes laser source, operating at the wavelength of 1053 nm. A pulse duration of 675 fs is estimated from the autocorrelation trace measurement provided by a recurrent autocorrelator (PulseCheck, APE). The laser beam is Gaussian shaped and focused by means of a 650 mm focal length lens on the sample. The focused beam diameter is about 155 μm at 1/e. For each pulse, the energy density which irradiates the sample is calculated by measuring the energy and the beam diameter in sampling paths, as shown in Fig. 1. The energy measured in the sampling path is calibrated with respect to the absolute energy value that irradiates the surface of the tested sample. The beam profile is recorded in a plane optically equivalent to that of the sample. The equivalence between these two planes, of main importance for the proposed method, has been verified and confirmed by analyzing the beam in both the sampling and the direct paths and by studying the effect of self-focusing in the direct paths. The calculation of the B-integral, which represents the accumulation of nonlinear phase along the beam propagation, has been done. In this study, the maximal energy used was 1.5 mJ. Assuming the hypothesis introduced by Nauport et al. [13], the simplified form of the B-integral leads to B ∼0.76 in our operating conditions. It is below the self-focusing limit which can be taken at B ∼1. Thus, the effect of self-focusing is nonexistent in this study and confirms the equivalence between the two paths. Given the error margin of each instrument, the energy density is determined with an absolute accuracy better than 10%.

To determine the 1/1 LIDT, we irradiate 10 independent sites per fluence and repeat the procedure for more than 10 fluences. A differential interferential contrast (DIC) microscope (Axio Imager.A2, Zeiss, with a 0.90 NA, 100 x objective) is used to observe the damage sites and determine the damage threshold of a sample, i.e., the highest fluence for which no damage event occurs. The result of a 1/1 test performed on a 200 nm silica single layer at an angle of incidence of 45° is reported in Fig. 2. The graph exhibits a steep slope, characteristic of 1/1 LIDT tests in short pulse regime. The LIDT is defined as the mean between the highest fluence for which no damage occurs and the lowest fluence for which the damage probability is different from zero. The uncertainty is set as the difference between this mean and these highest and lowest fluences. The LIDT is established at 5.37 ± 0.06 J/cm² for this silica single layer. The uncertainty can be reduced by testing more fluences near the damage probability transition. For a fluence slightly higher than 5.37 J/cm², we observe a surface modification, as shown in Fig. 2. Because of the deterministic behavior of damage process in this regime, one can determine easily and precisely the lowest fluence at which damage occurs with a probability of 100%.

For higher fluences of irradiation, the delamination of the damage area, shown in the zoomed-in Fig. 3(b), the presence of a slight surface modification similar to that on the damage site reported in Fig. 2 is observed. Considering a Gaussian-shaped beam, this similarity in morphologies is consistent with the fact that the edge of the damage site is irradiated with a lower fluence than the center.

Alternatively, another method is widely used in the ablation community to determine the damage threshold of a material. Based on microscopic observations, it consists of measuring the area of the damage site created at fluences higher than the damage threshold of the material and estimating the ablation threshold by extrapolation of the area to zero. Initially used to determine the size of a Gaussian spot [14], this method has

![Fig. 1.](image1.png)  
**Fig. 1.** Experimental characterization of the laser irradiations. The “half-wave plate + polarizer” system at the output of the laser is used to control the energy. The Gaussian-shaped profile reported here has been recorded in a plane equivalent to that of the sample.
been applied in ablation studies [15,16]. It is based on the use of a Gaussian spot: since the energy repartition in a Gaussian beam fits an exponential function, it results in a logarithm dependence of the damaged area on the fluence. This method has been carried out on the damage sites formed during the 1/1 test of the silica single layer previously exposed in Fig. 2. The LIDT is set by fitting damage surfaces by a logarithm function of the fluence and by extrapolating this fit to zero, as shown on Fig. 4. For each fluence bin, the mean area \( \mu_F \) and a dispersion set at \( 2\sigma_F \) are represented, \( \sigma_F \) being the standard deviation. The uncertainty on the LIDT has been determined with a Monte Carlo method: for each fluence bin, a random point is obtained from a uniform distribution defined over the interval \( [\mu_F - \sigma_F; \mu_F + \sigma_F] \). Then, a logarithm fit is done for these points and extrapolated to zero. This procedure is repeated several times: we chose twice the standard deviation on the intersection values to be the uncertainty on the LIDT. With this method, we set the LIDT at \( 5.50 \pm 0.09 \) J/cm\(^2\): it is in good agreement with the 1/1 test taking into account the error bars.

We now present the mono-shot method: this method consists of irradiating the component at a fluence much higher than its LIDT to create a well visible damage site, as the one on Fig. 3. For a pulse, the spatial profile of the beam and the energy of the pulse are recorded: therefore, the fluence of irradiation on the sample is calculated. Since the focused beam is Gaussian shaped, a tested site is irradiated by a fluence gradient which decreases from the center of the beam, where the fluence is maximum (\( F_{\text{max}} \)), to the edges. The knowledge of the energy repartition in the spot permits us to calculate the local fluence in each position of the beam. Consequently, with a unique shot, the tested area is illuminated with a continuous range of fluences, from 0 to \( F_{\text{max}} \) J/cm\(^2\). After damage is created, an observation of the damage morphology is performed by means of a DIC microscope. Image processing of these micrographs is illustrated in Fig. 5. The damaged area is encircled: for the silica layer example, as illustrated in Fig. 3, all the modified surface that can be observed with the same magnification as the one use for 1/1 damage discrimination is surrounding. As the test is performed at an angle of incidence of 45°, this area is elliptical. Thanks to the recording of the beam profile, the corresponding spatial profile of the laser beam is superposed and centered on the damage area. A correspondence between the edge of the damage area and local fluence in the beam is done: the local fluence at the edge of the damaged area is the minimum fluence necessary to trigger the damage; in other words, the LIDT of the material. In Fig. 5, we have applied this technique on three sites initiated at three different fluences. The LIDT is measured on the horizontal section of the ellipse; we read (a) 5.64 J/cm\(^2\), (b) 5.62 J/cm\(^2\), and (c) 5.36 J/cm\(^2\). On site (a), we have also inspected the local fluences correlated to the perimeter of the damage area: the measured LIDT is \( 5.68 \pm 0.20 \) J/cm\(^2\); the uncertainty setting at twice the standard deviation. For this site, the mono-shot method yields an error of about 3.5%, of the same order of magnitude, compared with conventional methods. On site (c), the fluence of test is slightly higher than the laser damage threshold of the material. The part of the beam that participates to the formation of the damage area corresponds to the center of the Gaussian, where the gradient in local fluence is close to zero. Thus, a small increase in the fluence leads to a strong increase in the damage area: this must
explain the high fluctuations in damage areas seen in Fig. 4 for fluence close to the laser damage threshold. However, because of the small gradient in local fluence, the evolution of the damage morphology from the center to the edge is smoother and the determination of the damage area is less precise. On sites (a) and (b), the edges of the damage area have been irradiated by the tails of the Gaussian beam, where the gradient in fluence is higher: transition in morphology is clearer. For that reason, it seems better to use a fluence of irradiation clearly higher than the 1/1 LIDT.

Comparing the LIDT assessed with the 1/1 test, 5.37 ± 0.06 J/cm², results obtained with the mono-shot method are relevant, given the error bars. The contrast of the micrograph, the lighting, and the damage morphology (nature of surface modifications) are the major sources of error in the assessment of the mono-shot LIDT; they can bias the delimitation on the damaged area. An inhomogeneity of the coating can also introduce dispersion on the LIDT values, and vice versa: the uncertainty in the measure can bring information on the homogeneity of the coating at a micrometer scale by analyzing the damage morphology, or at the centimeter scale by performing mono-shot measurements at different locations. Other samples have been tested with this method. For each sample, two sites are tested with the mono-shot method. Results are reported in Table 1 and compared to the LIDT values obtained with the 1/1 procedure. The analysis of these data shows that the use of a unique shot gives access to the LIDT of materials with a difference between standard procedure and the mono-shot procedure on average equal to 2.5% and lower than 6%. Moreover, one can note that this method is applicable for a large set of optical components, showing an interest for many applications. Thus, the mono-shot technique provides a good first estimation of the properties of an optical component.

This fast and simple method can be used for the determination of the LIDT of a component, intrinsic to the electronic property of materials as opposed to defect-induced damage. By performing a large set of measurements on dielectric coatings, we have shown that it is possible to determine the LIDT of a material after one irradiation in short pulse regime with a unique laser shot and a good reliability. The main advantage of this method lies in the saving on tested surface and on time spent: this principle offers a simple straightforward method using few hundreds of square micrometers sufficient to estimate the LIDT of a material. It has been carried out on a large variety of dielectric components, and the LIDT results exhibit a small difference, lower than 6%, from the results obtained with the standard method. As another advantage, considering that the beam profile is recorded and correlated to the damage morphology, this method allows theoretically the use of a non-Gaussian spot. Thus, it is applicable to many experimental facilities. Moreover, in the context of expensive or unique optical components or of the prospect for new dielectric materials scarce and difficult to coat, such a mono-shot test could be performed on a edge of an optic to estimate its LIDT and thus ensure a safe use of the laser system.

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**REFERENCES**


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**Table 1. Comparison of LIDT Measured with the Mono-Shot Method and the Standard 1/1 Test Procedure**

<table>
<thead>
<tr>
<th>Sample</th>
<th>Deposition Process</th>
<th>Angle of Incidence (°)</th>
<th>Mono-Shot LIDT (J/cm²) (2 sites per sample)</th>
<th>1/1 LIDT (J/cm²)</th>
<th>Absolute Difference between 1/1 and m-s LIDT (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>HfO₂ single layer</td>
<td>E-beam + IAD</td>
<td>45</td>
<td>5.19</td>
<td>5.43</td>
<td>4.4</td>
</tr>
<tr>
<td>Sc₂O₃ single layer</td>
<td>IBS</td>
<td>15</td>
<td>5.02</td>
<td>4.91</td>
<td>2.4</td>
</tr>
<tr>
<td>Quarter-wave mirror</td>
<td>E-beam</td>
<td>45</td>
<td>5.01</td>
<td>2.93</td>
<td>2.0</td>
</tr>
<tr>
<td>Broad bandwidth mirror</td>
<td>E-beam + IAD</td>
<td>0</td>
<td>1.34</td>
<td>1.32</td>
<td>0.8</td>
</tr>
<tr>
<td></td>
<td></td>
<td>0</td>
<td>1.06</td>
<td>1.07</td>
<td>0.9</td>
</tr>
<tr>
<td></td>
<td></td>
<td>40</td>
<td>1.11</td>
<td></td>
<td>3.7</td>
</tr>
</tbody>
</table>

**Absolute Difference between 1/1 and m-s LIDT (%)**

0.06 J/cm² corresponds to the 1/1 LIDT.