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Discussing defects related to nanosecond fatigue laser damage: a short review

Frank Wagner,* Alexandre Beaudier, and Jean-Yves Natoli

Aix Marseille Univ, CNRS, Centrale Marseille, Institut Fresnel, Marseille, France

Abstract. Laser damage measurements with multiple pulses at constant fluence (S-on-1 measurements) are of high practical importance for design and validation of high-power photonic instruments. Using nanosecond lasers, it was recognized long ago that single-pulse laser damage is linked to fabrication-related defects. Models describing the laser damage probability as the probability of encounter between the high fluence region of the laser beam and the fabrication-related defects are thus widely used. Nanosecond S-on-1 tests often reveal the "fatigue effect," i.e., a decrease of the laser damage threshold with increasing pulse number. Most authors attribute this effect to cumulative material modifications operated by the incubation pulses. We discuss the different situations that are observed upon nanosecond S-on-1 measurements that are reported in literature and speak in particular about the defects involved in the laser damage mechanism. These defects may be fabrication related or laser induced, stable or evolutive, cumulative or of short lifetime. @ 2018 Society of Photo-Optical Instrumentation Engineers (SPIE) [DOI: 10.1117/1.OE.57.12.121904]

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1 Introduction

To make meaningful laser damage tests that do not require huge budgets, one usually chooses the S-on-1 protocol.¹ The S-on-1 protocol uses a high number of pulses (*S* pulses) of constant fluence on each test site mimicking the operation of a typical high-power photonics setup or instrument, which is expected to work with constant performance over a long time. However, comparing the S-on-1 damage threshold T(S) to the single-pulse (1-on-1) damage threshold,² one often recognizes that the damage threshold decreases for increasing pulse number *S*. This effect is often called "fatigue effect."³

Independent of the aforementioned observation, it is true that for nanosecond lasers, the damage thresholds are typically smaller than the damage threshold that is expected from the perfect optical material, and the damage morphology close to the threshold consists of many small craters.^{4–6} In consequence, laser damage is attributed to defects in the host material.^{7,8}

In this review paper, we want to discuss what kinds of defects may be related to the fatigue effect at different wavelengths basing ourselves on a variety of already published data.⁹

2 Material Modification Fatigue and Statistical Fatigue

The name "fatigue effect" has been chosen in analogy with mechanics, where a metallic work piece that is charged and uncharged repeatedly can break after many cycles, even when using a load that is much smaller than the load it can withstand for a single loading/unloading cycle (Fig. 1). In fact, many laser damage T(S) plots look exactly like the plot in Fig. 1(a).^{10,11}

In mechanics, there is in fact a small crack developing slowly over thousands of cycles but then the sample suddenly breaks. It is thus natural that usually people first think of cumulative material modifications as the reason for the laser damage fatigue effect too.

At infrared wavelengths, however, the damage probability as function of pulse number P(S) often follows a simple statistical model that assumes a constant single-pulse damage probability (and thus does not consider cumulative material modifications).^{13,14} By the way, this statistical model has first been proposed in the early days of laser damage research by Bass and Barrett¹⁵ and was abandoned later on (for a discussion, see Refs. 3 and 13).

When working with longitudinal multimode nanosecond lasers, the irregular intensity spikes can explain this constant damage probability,¹⁶ but good agreement with the statistical model was also found for an experiment with a longitudinal monomode laser, for which the monomode operation was verified for each pulse.¹⁴

In the latter case, at least one step in the damage initiation process has to be statistical in nature, and in terms of the damage precursor model, one would say that we deal with light-induced transient damage precursors of short lifetime. For potassium titanyl phosphate crystals, in particular, a damage mechanism model that is compatible with statistical fatigue has been reported.^{17,18}

One of the first takeaways of this paper should thus be that every fatigue effect [T(S)-decrease] is not due to cumulative material modifications (Fig. 2).

3 Material Modification Fatigue

Material modification fatigue is easily recognizable in the P(S) plots too, as they start in this case with a number of incubation pulses where the damage probability is zero.¹⁴ Typically, this case is encountered when using UV wavelengths. More generally speaking, statistical fatigue is frequent if the laser-matter interaction is weak, whereas

^{*}Address all correspondence to: Frank Wagner, E-mail: frank.wagner@ fresnel.fr



Fig. 1 Information on mechanical fatigue damage. (a) Mechanical fatigue damage in steel, according to Ref. 12. A chosen stress amplitude is applied and removed for a high number of cycles. During the first cycles, a small crack is initiated. The crack then grows slowly until the work piece beaks suddenly. (b) Photograph of an aluminum part that was subject to fatigue failure.¹³

material modification fatigue is frequent if the laser-matter interaction is strong.

In the following, we will discuss and evaluate different possible models for material modification fatigue.

3.1 Precursor Encounter Models and S-on-1 Tests

For understanding nanosecond laser damage, one needs to consider that even sparse defects in the optical material were greatly supported by the success of the precursor encounter models to describe variations in the 1-on-1 damage probability observed when changing the laser beam size. These precursor encounter models consider that the material can be described like a host material with homogeneously distributed defects inside and laser damage appears every time when a defect that may induce laser damage (a laser damage precursor) is irradiated by a fluence above its threshold.⁷

Having a good model for 1-on-1 laser damage at hand makes it natural to ask ourselves if we could not use this model for fatigue damage too. For an ideally stable laser, the answer to this question is clearly: no. The reason is simply that all precursor presence models suppose that the damage probability is the probability of encounter between the laser beam and a damage precursor. However, during an S-on-1 test that modifies the damage precursors (e.g., reducing their damage threshold), this process of encounter is no longer probabilistic and in consequence these models can no longer be applied.

Taking into account laser fluctuations, stable precursors (no material modifications) can explain some of the observed damage threshold decrease in S-on-1 tests.^{13,19} The limited pointing stability and pulse energy stability cause an increase of the "surface over threshold" that has been sampled during irradiation with pulse number 1 to *S*. According to this model, the damage probability increases significantly with increasing pulse number if the peak fluence in the beam is close to the damage precursor threshold $T_{\rm P}$. It, however, fails to explain the experimentally observed increase in damage probability for fluences that clearly exceed $T_{\rm P}$.¹³

As a conclusion to this section, we can say that precursor encounter models are not adapted to describe material modification damage.

3.2 Deterministic Material Modification Damage

The frequently reported observation that the slope of S-on-1 damage probability curves steepens with increasing S is thus

not an indication of increasing damage precursor density but denotes that multipulse damage becomes more and more deterministic with increasing pulse number because the material under the beam is modified in a deterministic way. This is true even for longitudinal multimode laser with energy fluctuations. We will now address the question whether it is rather the fabrication defects or the host material that is modified in a way to eventually cause fatigue damage.

3.2.1 Modification of fabrication defects

First, let us assume that the fabrication defects that form the damage precursor ensemble in the 1-on-1 laser damage model will be modified by the irradiation such that they will cause multipulse damage after a certain number of incubation pulses. Damage density measurements carried out during the development of the laser damage metrology for the inertial confinement fusion class lasers showed us that a power law is a good approach for the distribution of the precursor thresholds in typical optical materials.^{20,21} A shape close to a power law has also been obtained by thermal calculations starting from a size distribution of the defects as observed for metal clusters in solid materials.²² The powerlaw-like distribution of the damage precursor density as function of fluence means that we have lots of damage precursors under the beam. These defects, however, do not trigger laser damage during the first pulses because their threshold is larger than the applied laser fluence. We will now try to imagine that their damage threshold is lowered by the incubation pulses until they can trigger laser damage.

The defects that we are looking for are thus irradiated by the incubation pulses and do not react catastrophically to this level of irradiation. Limiting this discussion to damage precursors in the bulk, the main "suspects" of being laser damage precursors are submicronic metallic inclusions,²³ submicronic inclusions of nonstoichiometric dielectrics,²⁴ and clusters of point defects (such as color centers).²⁵ Some of these defects are accompanied by strain in their vicinity, which could lead to mechanical failure.²⁶

As the irradiation definitely has an effect on the material, it either generates electron-hole pairs (if absorbed in a dielectric) or/and causes heating of the conduction band electrons (if absorbed in a metallic inclusion). Most of these conduction band electrons will relax nonradiatively generating "moderate" heating. We speak about moderate heating, in the sense that no laser damage is caused by the pulse we



Fig. 2 Schematical summary of literature data. For references, please see the text. Dashed lines show expected dependencies for perfect lasers. Dots show typical data scattering for real lasers. (a) In case of a decreasing S-on-1 laser damage threshold *T* with increasing pulse number *S* (using otherwise identical measurement conditions), one speaks of a fatigue effect. (b) The damage probability *P* at constant fluence but varying pulse number *S* may show this shape that indicates statistical fatigue of the shape in (c), which indicates material modification fatigue. (d) For material modification fatigue, the obtained multipulse damage probability for larger test-beam sizes indicate an influence of self-focusing in the damage mechanism. (e) Shows all damaged sites in the fluence—shot number plane. Using the adapted beam size, one can observe "immediate" damage induced by fabrication defects as well as fatigue damage occuring for higher pulse numbers.

are looking at. (Extreme heating would lead to thermal runaway^{27,28} or strong thermal stresses, which would lead to damage during this pulse.) Moderate heating, which is not triggering damage, will either be without any influence to the material or favor diffusion (of metallic inclusions),²⁹ annealing of point defects,³⁰ and resorption of frozen stresses. In one word, the thermal compound of the relaxation rather leads to "laser annealing" than worsening of the damage precursors. We tried to depict these possible lightinduced material modifications in the schematics of Fig. 3.

The only mechanism that may worsen existing damage precursors concerns point defects. Point defects generate



Fig. 3 Possible modifications of pre-existing fabrication defects during the incubation pulses.

stresses in their vicinity^{26,31} and stresses favor the generation of point defects during the relaxation of electron–hole pairs.^{26,32} Additionally, point defects easily provide conduction band electrons and the presence of conduction band electrons early in the laser pulse will favor absorption and thus laser damage.¹⁸

Hence, for defect clusters, a worsening by the incubation pulses cannot be completely excluded but a well-studied crystal, KDP, gives a counterexample. Damage precursors in KDP are believed to be defects clusters;²⁵ however, KDP shows strong laser conditioning.³³ Hence, defect clusters too do not always worsen during preirradiation with nondamaging fluences.

In summary, it seems probable that fabrication defects (the single-pulse damage precursors) do not worsen during the incubation pulses of an S-on-1 test.

3.2.2 Modification of the host material

Second and last, we should thus look at the possibility that new defects are created in the host material and these new defects cause multipulse laser damage at fluence levels lower than the 1-on-1 threshold.

The creation of new defects by low fluence irradiation has been reported in several different materials, especially if the photon energy of the laser approached the bandgap of the material. More precisely, cumulative material modifications during the incubation pluses have been detected by modifications in the transient refractive index change in silicate glasses,³⁴ by changes in the (transient) photoacoustic signal in alkali-halide crystals,²⁶ by long living UV-absorption changes in fused silica fibers,³⁵ and, more recently, by long living changes in the Raman spectrum³⁶ as well as by changes of intermediate lifetime in the fluorescence intensity³⁷ and the absorption spectrum.³⁸ But, even if experimental evidence for cumulative material modifications is found, the link of these modifications with fatigue laser damage may be quite indirect³⁷ and thus rather difficult to prove. Long and detailed investigations, including the development of quantitative models, are necessary to "prove" the connection between the observed material modifications and the occurrence of laser damage. Two groups of references should be mentioned in this context (Refs. 26 and 34 and references therein) but others are also reviewed in Ref. 3.

Recent work on fatigue laser damage in synthetic fused silica using UV wavelengths indicates the importance of self-focusing for the damage mechanism.³⁹ Similarly, one of the most complete investigations on nanosecond laser damage also includes self-focusing as the final step in their model.³⁴ More generally, the light-induced modifications of the material may in fact modify the propagation of the laser beam by different physical mechanisms (modification of the refractive index, thermal self-focusing, and nonlinear self-focusing) and thus cause a catastrophic enhancement of the peak fluence a bit further downstream compared to the location of the light-induced defects. A signature of this first type of link between material modifications and laser damage can be obtained when working with lasers having a spatially Gaussian beam profile and a good positional stability. In this case, an influence of the beam diameter on the fatigue strength, i.e., the relative threshold decrease observed at a certain number of pulses [for example, T(1000)/T(1)], is observed.³⁴

A direct link between laser-induced defects and the occurrence of laser damage, where the light-induced defects absorb themselves a sufficient portion of the light to induce thermal run-away and damage, has in fact never been validated by a quantitative model.

A third idea of linking laser-induced defects and laser damage relies on mechanical strain in the vicinity of laser-induced color centers. At sufficient concentration of color centers, the strong strain could then cause nanometric cracks that join to form cracks large enough to cause field enhancement and finally laser damage at the location of the light-induced defects.²⁶ Contrary to the link by self-focusing, this scenario lacks supporting observations by other authors.

Summarizing Sec. 3.2, we might say that material modification fatigue is most likely to be caused by generation of new defects in the host material. These defects have long lifetimes and accumulate pulse after pulse until the effects they have on the laser beam become sufficiently strong to cause damage. Experimental observations point toward defectinduced self-focusing of the laser beam causing damage a bit further downstream^{34,39,40} rather than the defects absorbing themselves a sufficient portion of the light to induce thermal run-away and damage or the defects causing mechanical damage due to local strain in their vicinity.

4 Simultaneous Failure Modes During S-on-1 Tests

Even if the fabrication defects that act as laser damage precursors during single-pulse tests do not play an important role for fatigue damage, they are nevertheless there and may cause damage during the first pulses. Depending on the ratio between the beam diameter and the average distance between the 1-on-1 damage precursors, this failure mode can be dominating, thus hiding a possible fatigue effect that will show up once the material has been improved by decreasing its density of fabrication defects. For example, testing coatings in the UV with "large" beams did not show a fatigue effect.⁴¹

When using smaller beams, and thus decreasing the chance to hit a 1-on-1 damage precursor, one recognizes that the sites that did not damage during the first pulses due to fabrication defects are damaging due to a fatigue mechanism. In some cases, one can see the two failure modes (fabrication defect damage and material modification fatigue damage) in a single experiment.⁴¹

Further, considering that laser conditioning affects the 1-on-1 damage precursors, one could even say that "fatigue" and "conditioning" may be present in one and the same sample as these two effects are based on two different types of defects and two different types of physical mechanisms leading to damage.⁹

5 Summary and Conclusions

In summary, we provided a review of nanosecond multiple pulse measurements and discussed different empirical models for these experiments. We concluded that precursor encounter models should not be used to describe S-on-1 measurements showing material modification fatigue. The light-induced material modifications that finally lead to damage are probably located in the host material and not in the pre-existing fabrication defects that act as damage precursors for single-pulse tests. The preceding sentence should not be misunderstood in the sense that fatigue damage is

independent of the fabrication process. Different fabrication processes may very well generate host materials in which new defects can be created more or less easily, which induces different resistance to fatigue laser damage. The defects leading to material modification fatigue are light induced and of long lifetime so that they accumulate during the incubation pulses. We also showed that for weak laser-matter interaction, such as frequently encountered at IR wavelengths, statistical fatigue is frequently observed. The defects associated with statistical fatigue are light induced and of short lifetime so that they do not accumulate.

To distinguish between material modification fatigue and statistical fatigue, one should plot the damage probability P as function of maximum pulse number S. For statistical fatigue, these data are well described by the statistical model, and for material modification fatigue, the incubation pulses (with P = 0) are followed by a quick transition from P = 0 to P = 1.

The $N_{\rm d}(F)$ scatter plots of the damaged sites allow us to quickly understand if there are two parallel failure modes. For example, for some sites, fabrication defects may cause damage during the first pulses and for the sites without fabrication defects, intrinsic light-host material interaction may cause material modification fatigue causing damage at higher pulse numbers.

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Biographies for the authors are not available.