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Influence of the multilayer dielectric mirror design on the laser damage growth in the sub-picosecond regime

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The peak-power on high-power laser facilities is limited by the laser-induced damage on the final optical components. Also, when a damage site is generated, the damage growth phenomenon limits the lifetime of the component. Many studies have been performed to improve the laser-induced damage threshold of these components. The question now arises as to whether an improvement of the initiation threshold leads to a reduction of the damage growth phenomenon. To address this question, we performed damage growth experiments on three different multilayer dielectric mirror designs exhibiting different damage thresholds. We used a classical quarter-wave designs and optimized designs. The experiments were carried out with a spatially top-hat beam, spectrally centered at 1053 nm with a pulse duration of 0.8 ps in *s*- and *p*-polarization. The results evidenced the impact of design on the improvement of the damage growth thresholds and a reduction of the damage growth rates. A numerical model was used to simulate damage growth sequences. The results reveal similar trends to those observed experimentally. On the basis of these three cases, we have shown that an improvement of the initiation threshold through a modification of the mirror design can lead to a reduction of the damage growth phenomenon.

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1. INTRODUCTION

The peak power of petawatt-class lasers has steadily increased 2 since the development of the Chirped Pulse Amplification (CPA) 3 technique [1]. However, the peak-power remains limited by the laser-induced damage on the final optical components of 5 these facilities [2]. Laser-induced damage is driven by non-linear 6 photo-ionizations in the sub-picosecond regime. These phenom-7 ena lead to the critical electron density of the material which 8 triggers the damage mechanism [3–6]. The mechanisms imply 9 that the damage threshold is linked to the intrinsic properties of 10 the material, notably the bandgap energy [7, 8]. Several studies 11 have improved the understanding of damage mechanisms on 12 Multilayer Dielectric (MLD) coatings which depend on different 13 laser parameters such as the pulse duration for example [9–11]. 14

Laser-Induced Damage Threshold (LIDT) improvement of optical components of high-power laser facilities is not sufficient to avoid the occurrence of damage sites because of defects due to manufacturing process and of particle contamination [12–17]. The initial size of these damage sites is generally micrometric which is too small to have detrimental effects on the beam propagation. However, a significant increase of the damaged area has been reported after successive shots [18–20]. The determination the growth threshold and the damage growth rate is an important issue to predict the lifetime of these components.

In the sub-picosecond regime, laser-induced damage has a deterministic behavior, and LIDT depends on the Electric Field Intensity (EFI) distribution in the MLD stack [21-23]. To improve LIDT, different methods were explored such as a selection of materials with the highest intrinsic LIDT and/or an optimization of the stack design. Apfel et al. demonstrated an improvement of the LIDT by shifting EFI peaks into the most resistant material [24]. Previous works evidenced that optimizing only the outer layers can lead to a significant improvement of LIDT [25-27]. It was also reported that some designs could be very sensitive to manufacturing errors. Chorel et al. presented a Robust-Optimization Algorithm ROA of MLD coatings by taking into account manufacturing errors [28]. An improvement of the LIDT has been demonstrated by depositing three designs. The efficiency of the algorithm has been validated experimentally with an increase of approximately 44% between one classical design based on Quarter-Wave Optical Thickness (QWOT) and an optimized design based on the ROA [29]. As a follow-up to this study, the questions we want to answer are:



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Fig. 1. Representation of the DERIC test bench.

does an increase of LIDT lead to an increase of the growth threshold and do MLD designs have an influence on damage growth 45 rates? 46

47 To continue with the experimental validation of ROA, we 48 investigate the improvement of Laser-Induced Damage Growth 49 Threshold (LIDGT) along with the initiation threshold. In this paper, we present the experimental damage test bench used for 50 the measurement of initiation threshold and damage growth 51 experiments. Then, the designs of the three mirror samples are 52 detailed and the results of damage growth experiments are pre-53 sented. We used a spatially top-hat beam to illuminate damage 54 sites initiated intrinsically. LIDGT and damage growth coeffi-55 cients were measured to compare the damage growth dynamics 56 of the three samples. Finally, a numerical model in 2D with 57 the Finite Element Method (FEM) was developed to assess the 58 trends observed experimentally. 59

2. EXPERIMENTAL SET-UP 60

The experimental study was performed on the laser test bench 61 100 called DERIC at CEA-CESTA using a S-pulse Amplitude Sys-62 101 tèmes Laser source. This laser operates at 1053 nm and delivers 2 63 102 mJ pulses at 10 Hz. The pulse duration was chosen to be as short 64 as possible around 800 fs, estimated from the measurement of a 65 recurrent autocorrelator [30, 31]. On this facility, two different 66 and separated measurement beam-lines have been developed. 67 A scheme of the set-up and of the two beam-lines is reported in 68 Fig.1. 69

First, a spatially Gaussian beam is used to determine precisely 70 the intrinsic LIDT of the optical components. Second, a square 71 and top-hat beam is used to perform experimental growth mea-72 surements. A retractable mirror makes it easy to switch from 73 one beam-line to another. Regarding LIDT measurements, the 1on-1 procedure was used to assess initiation damage thresholds, 75 intrinsic to the structure and the materials, and to discriminate 76 which mirror exhibits the highest damage threshold [30]. In the 77 sample plane, the Gaussian shaped focused spot has a 160 μ m 78 diameter at 1/e. On this beam-line, taking into account the er-79

ror margin of each instrument, absolute fluence is given with an accuracy better than 10%. Damage detection is firstly estimated *in-situ* by means of a probe beam coupled to a Schlieren technique. Next, the occurrence of the damage is checked and validated post-mortem by means of a differential interference contrast (DIC) microscope (50× magnification) as recommended by the ISO standard [32]. This measurement beam-line is chosen to precisely measure the LIDT because the Gaussian spatial beam profile makes it possible to accurately determine the maximum beam fluence which is attributed to each laser irradiation. On the other hand, to perform accurate damage growth measurements, a homogeneous energy distribution deposited on the damage site is preferable [33]. In this way, analysis of the results is facilitated by decorrelating the laser damage dynamics from the fluence distribution of the beam. Thus, growth experiments have been carried out with a spatially square and top-hat beam. This profile is obtained by shaping the spatially Gaussian beam from the laser source with a silica phase plate associated with a 300 mm focal lens. This leads to a 150 μ m squared top-hat beam at a specific position along the beam propagation axis. With the maximum energy available on the sample around 2.0 mJ, the maximum fluence is around 8.0 J/cm^2 at the plateau of the top-hat beam. The process used for the damage growth experiments has been first described in [34]. In a few words, a high peak fluence profile is first used to trigger initial damage on the dielectric mirror. Then the sample is moved along the propagation axis to the top-hat position and the growth of this initial damage site is realized by irradiating it with the top-hat profile. A microscope coupled to a camera enables both observations, the growth sequence and recording of the whole sequence. The camera is synchronized with the laser shot repetition frequency to capture, shot by shot, the evolution of the damaged area. Thanks to the microscope acquisitions, damaged areas are estimated through image processing (using Image J software) that allows to determine the growth damage rate of each damage site.

3. CHARACTERISTICS OF THE THREE DEPOSITED DE- 178 116 SIGNS 117

In this study, three different mirrors have been studied. These 118 mirrors were developed during a previous study to assess the 119 influence of the thickness of some layers of the MLD stack to 120 improve the LIDT of the mirrors [28]. 12

Indeed, as described in the introduction of this paper, the 122 LIDT of MLD coatings subjected to sub-picosecond laser pulses 123 depends on the intrinsic laser damage threshold of the materials 124 (LIDT_{int}) and the Electric Field Intensity (EFI) distribution in the 125 stack [8]. The EFI is defined as the square norm of the electric 126 field normalized by the square norm of the incident electric field: 127 $EFI = |E|^2 / |E_0|^2$. Laser induced-damage will initiate where 128 the experimental fluence modulated by the EFI is higher than 129 the intrinsic LIDT of the material. Consequently, the LIDT of a 130 layer can be defined as : $LIDT_{layer} = LIDT_{int} \times EFI_{max}$ where 13 EFImax represents the maximum EFI value in the layer. Then the 132 180 LIDT of a multilayer is defined as the minimal LIDT value of its 133 181 constituent layers. With this description, one can see that a way 134 182 to increase the LIDT of mirrors is to modify the stack structure 135 183 in order to adjust the *EFI_{max}*. 136

184 The mirror design is composed of an odd number of QWOT 137 185 layers starting and finishing with the high refractive index ma-138 terial ([HL]ⁿ H). Having the high index material as outer layer 139 compared to low index material increases the refractive index 140 contrast of the first interface of the mirror with the incident 141 medium, air. Higher the index contrast induces higher reflectiv-142 ity of this interface consequently less electric field is propagating 143 in the stack. Moreover, thick low index overcoat (2L overcoat) usually used on ns high power laser facilities transport mirrors 145 193 are hosting high EFI peak value. In the present context, it is then 146 interesting to remove this overcoat layer. The EFI of a [HL]ⁿ H 147 design has its highest value at the interface between the two 194 148 outer layers of the MLD stack. Apfel has shown that by modify-149 ing the thicknesses of those two outer layers, the EFI peak could 150 be moved into the most resistant of the two materials which 151 should increase the LIDT [24]. This shift is induced by increas-152 ing the thickness of the low refractive index and most resistive 153 layer, and decreasing the thickness of the other layer. Based 154 on this consideration, Chorel et al. have developed a Robust 155 Optimization Algorithm (ROA) that modifies several layers and 156 takes into account the manufacturing errors [28]. The number 157 of layers to optimize is set as an input parameter of the method. 158 Other input parameters of the ROA are the refractive indexes 159 and the intrinsic LIDT of the materials involved in the stack. 160 Experimental validation of this ROA method has previously 16 shown an improvement of 44% of the LIDT [29]. 162

To investigate the influence of coating mirror designs and 163 the improvement of the LIDT on the laser damage growth phe-164 nomenon, we performed damage growth characterization on 165 three different designs counting the same odd number of layers 166 (37), all composed of hafnia and silica layers deposited on identi-167 cal high damage threshold BK7 substrates by unassisted electron 195 168 beam evaporation and in the same coater. The first design called 169 196 **QWOT** is a mirror composed of quarter-wave optical thickness 170 197 layers. The second design called **AOD** is inspired by one of 17 198 the Apfel approaches described above, where the thicknesses of 172 two outer layers are modified and weighted (balanced) by the 200 173 intrinsic LIDT of the materials. The last design called **ROAD**, 201 174 calculated with the ROA method has its 12 outer layers modified. 202 175 All 3 mirrors are designed to provide a reflectivity higher than 203 176 99% for s and p-polarized beam at 1053 nm with an angle of 204 177

incidence of 45° and for ambient air.

4. LIDT MEASUREMENTS

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Designs	<i>p</i> -polarization		s-polarization	
	EFI	LIDT (J/cm ²)	EFI	LIDT (J/cm ²)
QWOT	1.03	2.73 ± 0.20	0.70	4.36 ± 0.31
AOD	0.85	3.83 ± 0.18	0.49	6.12 ± 0.11
ROAD	0.67	3.84 ± 0.43	0.43	6.12 ± 0.16

Table 1. Results of 1-on-1 damage tests performed on the three mirror designs.

First, the intrinsic LIDT of each material deposited as monolayer was measured with 1-on1 tests at normal incidence. The intrinsic LIDT values were determined at 6.23 ± 0.13 J/cm² and $3.08 \pm 0.10 \text{ J/cm}^2$ for SiO₂ and HfO₂ respectively. Then, the LIDT values of all three designs were measured, as exposed in section 2, on the "Gaussian" beam-line using the 1-on-1 procedure at 45° of incidence, 0.8 ps of pulse duration, in ambient air for *s*- and *p*-polarization. Results are reported in Tab.1. Similar LIDTs were measured for both polarizations, on optics coated with the AOD and the ROAD. Lower EFI values induce higher LIDTs in *s*-polarization than in *p*-polarization. Those LIDTs are approximately 40% higher than the LIDT values of the QWOT design which confirms that optimizing the thicknesses of the outer layers increases the LIDT.

5. DAMAGE GROWTH EXPERIMENTS



Fig. 2. Evolution of the damaged area as a function of the number of shots. Three sites have been illuminated under the same conditions at a fluence of 2.84 J/cm^2 in *s*-polarization. The experiments were carried out on the AOD sample.

Laser induced-damage growth experiments were performed on the "Top-hat" beam-line. First, initial damage sites were created intrinsically with a Gaussian beam at a fluence 10% above the LIDT (with a single shot). The initial damage sites have an elliptical form with a major axis diameter between 25 and 30 μ m (see Fig.3(#1)). Those damage sites were illuminated with successive laser shots with the top-hat beam at different fluences (see Fig.2 and Fig.3). For a given fluence, 3 different sites were illuminated under the same conditions. The evolution of the area of the damage sites, as illustrated in Fig.2, revealed a



Fig. 3. Microscope acquisitions of the evolution of the damaged area. Damage growth experiments were performed on (a) QWOT design, (b) AOD, and (c) ROAD at a fluence of approximative 3.41 J/cm² in *s*-polarization. See Visualization 1 for complete sequences.

205 good repeatability of the damage growth phenomenon between206 different sites illuminated under the same conditions.

The results evidence three growth regimes: an initial regime 207 with a limited evolution of the size of the damage site, a main 208 regime with a linear evolution of damaged area, and a saturation 209 regime which corresponds to a decrease of the damage growth 210 until stabilization of the size of the damage sites. These three 211 regimes were described in ref.[33]. In this case, the duration of 212 the initial regime (number of shots) was very short for the three 213 214 mirror designs (around 50 shots).Damage growth coefficients were measured by only considering the main regime, described 215 with the following affine relationship [34–36]: 216

$$S_N = \beta \times N + S_0. \tag{1}$$

where S_N (μ m²) represents the damaged area after *N* shots, β (μ m²/shot) is the linear damage growth coefficient and S_0 (μ m²) represents the initial size of the damage site. For the 3 sites illuminated under the same conditions, the average between the 3 measured β coefficients is reported for each fluence.

The influence of the polarization on the damage growth phe-222 nomenon has been studied in ref.[37]. Damage growth thresh-223 olds (LIDGT), damage growth rate, and damage site morphology 224 have been shown to depend on polarization. Figure 4 shows 225 the evolution of the damage growth coefficients β as a function 226 of the fluence for the three different mirror designs. LIDGT is 227 defined as the average deviation between the highest fluence 228 with the growth coefficient β equal to 0 and the lowest fluence 229 with a non-zero growth coefficient. LIDGT values of the three 230 different designs are reported in Tab.2. 231

In the case of *p*-polarization, LIDGT for the QWOT design 232 is equal to $1.27 \pm 0.04 \text{ J/cm}^2$ and for the AOD is equal to 233 2.11 ± 0.01 J/cm². The highest LIDGT is found for the ROAD 234 at 2.35 ± 0.03 J/cm². This evidences an increase of the damage 235 growth threshold between the QWOT and the two optimized 236 designs, which can also be seen in Fig.4(a). It represents an in- 242 237 crease of approximately 66% and 85% of the LIDGT from QWOT 243 238 to AOD and QWOT to ROAD respectively. These improvements 244 239 are higher than that observed for the LIDT. However, the dam- 245 240 age growth rate seems similar for the three mirrors. 246 241



Fig. 4. Evolution of the damage growth coefficients as a function of the fluence for the three mirror designs for (a) *p*-polarization and (b) *s*-polarization. Vertical error bars represent the standard deviation between the three measured coefficients on different sites on the same sample under the same illumination conditions and fluence. The inset in figure (b) shows a zoom of the chart between 0.5 and 1.5 J/cm². Vertical dashed lines represent the LIDT of each mirror designs.

In the case of *s*-polarization, the results demonstrated also an improvement of the LIDGT as illustrated in the inset of Fig.4(b). We determined an improvement of approximately 29% and 35% of the LIDGT between QWOT-AOD and QWOT-ROAD respectively, which is lower compared to the improvement of the LIDT

(40%). The damage growth rate seems to be influenced by the 247 mirror design. Under low fluences, damage growth coefficients 248 for the three designs show values close to each other. Above 249 a fluence of 2.50 J/cm^2 , the damage growth coefficients of the 250 QWOT design are higher than for the two optimized designs. 251 Furthermore, the AOD has damage growth coefficients lower 252 than the ROAD. The increase of the LIDT leads to an improve-253

ment of the LIGDT that depends on the polarization 254

Designs	LIDGT (J/cm ²)		
Designs	<i>p</i> -polarization	s-polarization	
QWOT	1.27 ± 0.04	0.95 ± 0.03	
AOD	2.11 ± 0.01	1.23 ± 0.04	
ROAD	2.35 ± 0.03	1.28 ± 0.04	

Table 2. Measurements of damage growth thresholds on the three design mirrors.

6. MODELING OF THE DAMAGE GROWTH PHE-255 NOMENON 256

A 2D model using Finite Element Method (FEM) to solve the 25 Maxwell's equations was developed to simulate damage growth 258 sequences, as described in ref. [33]. The model is based on the 250 hypothesis of a growth phenomenon linked to the Electric Field 260 Intensity (EFI) in the MLD structure. The main goal of this model 26 is to assess the different trends observed experimentally. To 262 simulate damage mechanisms, the EFI distribution is calculated 263 with an initial damage site in the first layer of the mirror design. 264 Where the incident fluence modulated by the EFI reaches the 265 intrinsic LIDT of the material, the value of the refractive index 266 299 is replaced by the refractive index of air. The model is restarted 267 with the new damaged structure. 268

269 The three mirror designs were implemented in the model. The dimension of the geometry was set to $400 \times 9.6 \ \mu m^2$. The 400 270 *µ*m length corresponds to the field of view of the microscope dur-271 ing the experiments. The 9.6 μ m corresponds approximately to 272 the total thickness of the mirrors with 1 μ m of air and substrate 273 domain thickness. The geometry is meshed with a maximum 274 cell size equal to $\lambda/10$ (λ corresponds to 1053 nm). Damage 275 growth sequences at different fluences were performed only 276 in s-polarization since a 3D model is needed to simulate dam-27 278 age growth sequences in *p*-polarization [37]. Damage growth sequences are set to have 200 iterations. At the end of the it-279 erative process, the length of the damage site in the top layer 280 is measured, and compared to the damaged length observed 281 experimentally (see Fig.5(a)). The evolution of the length of the 282 damage site revealed similar trends as observed experimentally 283 with three growth regimes, illustrated in Fig.5(b). We consid-284 28 ered only the main regime to measure linear damage growth coefficients based on this relation [33]: 286

$$L_N = \alpha \times N + L_0. \tag{2}$$

 L_N (µm) represents the damaged length after N shots, α 28 $(\mu m/shot)$ is the linear damage growth coefficient and L_0 (μm) 303 288 represents the initial damaged length. 304 289

Figure 6 represents the damage growth coefficients calculated 305 290 numerically for each mirror design. The model reveals an im- 306 29 provement of the LIDGT between the QWOT design and the 307 292



Fig. 5. (a) Example of a numerical damage growth sequence on the AOD after 15 iterations with an incident fluence of 3.8 J/cm^2 in s-polarization. The color scale represents the intensity of the incident beam, the gray scale represents the refraction index. (b) Evolution of length of the damage site through the entire sequence for the three mirror designs at a fluence of 3.3 J/cm^2 . Only the first 80 of the 200 iterations are displayed.

optimized designs as observed experimentally. The model evidences different damage growth rates for the mirror designs (see Fig.5(b)). A stronger damage growth rate was calculated for the QWOT design. In addition, lower damage growth coefficients were evidenced for the AOD than for the ROAD as observed experimentally. This result motivates to the study the influence of mirror designs on damage growth phenomenon more broadly. These trends revealed by the numerical calculations are consistent with the experimental observations.



Fig. 6. Evolution of numerical damage growth coefficients in *s*-polarization as a function of the fluence following the three mirror designs.

7. CONCLUSION

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The experimental results revealed an improvement of 40% of LIDT between the classical and the two optimized designs. Damage growth experiments were performed on these mirrors to measure the damage growth threshold in the two linear polarization states. The results showed an improvement of the growth

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threshold between 29% and 85% depending on the polarization 308 state. In the case of *s*-polarization, a reduction in damage growth 370 309 rate is evidenced as a function of mirror designs, particularly 371 310 372 between AOD and ROAD while similar initiation and growth 311 373 thresholds have been measured. These results reveal the influ-312 ence of the mirror design on the damage growth phenomenon. 313 375 A 2D FEM model was used to assess the trends observed exper-314 376 imentally. The numerical results demonstrated similar trends 315 to that observed experimentally with an improvement of the 378 316 LIDGT between the QWOT and the two optimized designs. The 379 317 numerical model was developed to validate experimental trends. 380 318 It cannot to be fully representative and completely accurate first, 381 319 because we cannot compare damaged length against damaged 382 320 383 area and second the numerical model takes into account only 321 384 the electric field intensity distribution. To improve the repre-322 385 323 sentativeness of the simulation, a 3D model including different laser damage process such as the incubation phenomenon and 324 the deposition of debris should be developed. In addition, lower 325 388 numerical damage growth coefficients were calculated with the 326 380 AOD than with the other two designs. To conclude, an improve-327 ment of the initiation threshold through a modification of the 328 391 mirror design can lead to a reduction of the damage growth 392 329 phenomenon. These results underline the possibility of an op-393 330 timization of MLD designs by taking into account the growth 394 331 395 phenomenon. 332

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342Data availability.Data underlying the results presented in this408343paper are not publicly available at this time but may be obtained from409344the authors upon reasonable request.410

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