LLNL-TR-716785

LAWRENCE
LIVERMORE
NATIONAL
LABORATORY

Pico-second laser materials interactions: mechanisms, material lifetime and performance optimization Ted Laurence(14-ERD-014)

T. Laurence

January 5, 2017

This work performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

Pico-second laser materials interactions: mechanisms, material lifetime and performance optimization

Ted A. Laurence (14-ERD-014)

Abstract

Laser-induced damage with ps pulse widths straddles the transition from intrinsic, multi-photon ionization- and avalanche ionization-based ablation with fs pulses to defect-dominated, thermal-based damage with ns pulses. We investigated the morphology and scaling of damage for commonly used silica and hafnia coatings as well as fused silica. Using carefully calibrated laser-induced damage experiments, *in situ* imaging, and high-resolution optical microscopy, atomic force microscopy, and scanning electron microscopy, we showed that defects play an important role in laser-induced damage for pulse durations as short as 1 ps. Three damage morphologies were observed: standard material ablation, ultra-high density pits, and isolated absorbers. For 10 ps and longer, the isolated absorbers limited the damage performance of the coating materials. We showed that damage resulting from the isolated absorbers grows dramatically with subsequent pulses for sufficient fluences. For hafnia coatings, we used electric field modeling and experiments to show that isolated absorbers near the surface were affected by the chemical environment (vacuum vs. air) for pulses as short as 10 ps. Coupled with the silica results, these results suggested that improvements in the performance in the 10 -60 ps range have not reached fundamental limits. These findings motivate new efforts, including a new SI LDRD in improving the laser-damage performance of multi-layer dielectric coatings. A damage test facility for ps pulses was developed and automated, and was used for testing production optics for ARC. The resulting software was transferred to other laser test facilities for fs pulses and multiple wavelengths with 30 ps pulses. Additionally, the LDRD supported the retention and promotion of an important staff scientist in high-resolution dynamic microscopy and laser-damage testing.

Background and Research Objectives

Laser-induced damage of optical materials is a particularly interesting example of a laser-material interaction, in which the material itself is modified, often in ways contrary to its intended use. Laser-induced damage for sub-picosecond (ps) pulses and for pulses 1 ns and longer have been studied extensively. Thermal processes due to extrinsic absorption by materials not native to the host (bulk) dominate laser-induced damage in the ns-scale regime [1]. For short enough laser pulses (fs regime) thermal processes no longer apply and multiphoton ionization and tunneling dominate. However, the physics governing the intermediate regime (from 1 ps-100 ps) where typical pulse intensities range between 100GW/cm^2 and 1TW/cm^2 remained unclear. Models which explain the transition from defect-driven thermal-based breakdown in the ns-regime to photo-ionization and avalanche ionization induced free electron breakdown in the fs-regime are lacking, and measurements have not focused on this transitional pulse width regime, with a few exceptions [2].

Uncertainty in the physics driving breakdown by pulses in the ps-regime greatly complicates current efforts to model and improve performance limits and lifetimes of optics used in ps laser systems. For example, advanced short pulse laser systems are typically limited in their final optics to <0.5 TW/cm^2 in the ps regime, making these systems inherently inefficient as well as large and costly. Additional studies focused on the mechanisms and materials involved in laser damage of ps-scale laser pulses were needed to help improve laser performance.

For short pulse lengths, multi-layer dielectric coatings are used to create reflective optics that avoid nonlinear propagation issues through optical materials. Commonly used materials include silica and hafnia deposited in layers. By alternating layers of the low index material (silica) and the high index material (hafnia), it is possible to create highly reflective mirrors with greatly improved damage performance. Determining if there are possible avenues to raise the fluence for the onset of damage formed the primary objective of this LDRD.

The goals of the proposed scope were to: 1) measure short pulse (1-60 ps) damage precursor density, and determine if intrinsic or extrinsic factors limit performance; (2) determine effects of multiple short pulses, including growth and long term degradation; (3) investigate mechanisms of short pulse laser damage, and measure key parameters for modeling; (4) identify dominant failure mechanisms and damage precursors, and explore mitigation strategies.

Scientific Approach and Accomplishments

We accomplished these goals using an experimentally-driven approach, coupling carefully calibrated laser-induced damage measurements with several high-resolution microscopic methods to understand the density and nature of the defects leading to damage. Modeling efforts for understanding damage scaling laws as a function of pulse width, extracting damage precursor densities, optical field intensities in multi-layer stacks and propagation of fields through bulk silica and air were critical to the interpretation of these results.

A main goal of this project was to determine whether the laser induced damage observed between 1 and 60 ps is due to intrinsic limitations or is due to extrinsic factors such as defects. We studied the most common materials used to make multi-layer reflective coatings in high power laser systems: silica and hafnia. We performed measurements on silica and hafnia coatings as well as bulk fused silica finished with the best surface finish. We used strategies developed for detecting defect densities in the defect-dominated ns pulse length regime, including matching the position of damage to local fluences [3], as well as damage tests with varying beam sizes [4]. For ps pulses on entrance surfaces of optics, the damage site sizes tend to be much smaller (50 nm – 5 µm) than corresponding damage on exit surfaces with ns laser pulses important for NIF-related studies. The small damage site size allows detection of defect-related densities much higher than found previously, but requires the use of high-resolution microscopic methods to quantify the damage densities.

The laser damage test experiments were performed at the ACL laboratory, which was being developed as a laser-damage test facility in support of the Advanced Radiographic Capability (ARC) on NIF. One beam-line in vacuum was already in development for ARC-related damage testing [5–7]. We developed a beam-line in air that allowed for greater flexibility in adjusting beam size and allowing for higher fluence tests. It also allowed *in situ* imaging of growth of laser-induced damage, and a wider range of angle-of-incidence measurements. Software for control of laser damage

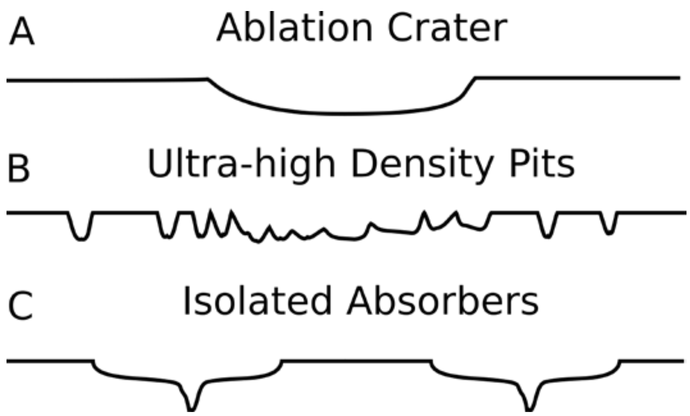

Figure 1: Schematics of damage morphologies found in laser-induced damage for 1-50 ps pulse widths. **A.** Smooth ablation crater. **B**. High density, individual pits that coalesce to form large damage craters. **C**. Isolated absorbers with small, deep pit associated with smooth, wide, shallow craters.

testing, correcting for beam energy and beam profiles, *in situ* monitoring of the damage process, and S/1 damage testing were developed and used in both beam-lines and transferred to other facilities.

We used high-resolution confocal optical microscopy, scanning electron microscopy, and atomic force microscopy to understand the nature of the resulting damage [8]. Damage classification, quantification of damage densities and pulse length scaling allowed us to understand several features about the physical mechanisms leading to damage in the different cases. The damage detected was classified into three distinct types shown in Figure 1: standard material ablation, ultra-high density pits, and isolated absorbers. The last is of particular importance in coating materials, as opposed to bulk fused silica (see Figure 2). The morphology of the observed damage, coupled with the reliable extraction of damage precursor densities, demonstrated that isolated defects causing damage to surrounding regions were limiting the damage performance of coatings with pulse widths longer than 1 ps, especially for those 10 ps and longer.

We modeled the laser-induced damage process of ablation using standard models [2,9], and compared these results with extracted scaling laws. The difficulties and inconsistencies in fitting these experimentally determined scaling laws for 1-60 ps add support to the finding that defect-driven damage is dominant in this pulse width range.

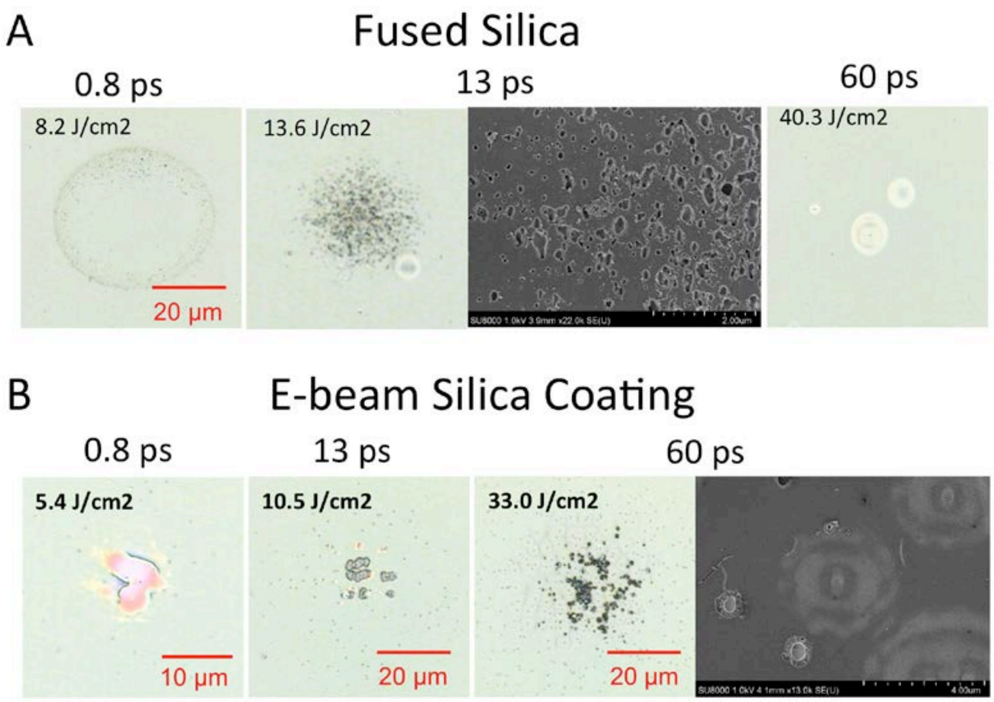

Figure 2: Confocal images and SEM images of basic types of damage found in fused silica and e-beam silica coatings. **A.** Smooth ablation is found for 0.8 ps pulses, high-density pits for 13 ps pulses, and isolated absorbers for 60 ps are observed in fused silica. **B.** Isolated absorbers dominate the laser-induced damage observed in silica coatings. For short pulses less than 10 ps, coating removal is most often observed. The image shown for 0.8 ps is for a transitional case where the coating is lifted off the substrate, but not removed entirely.

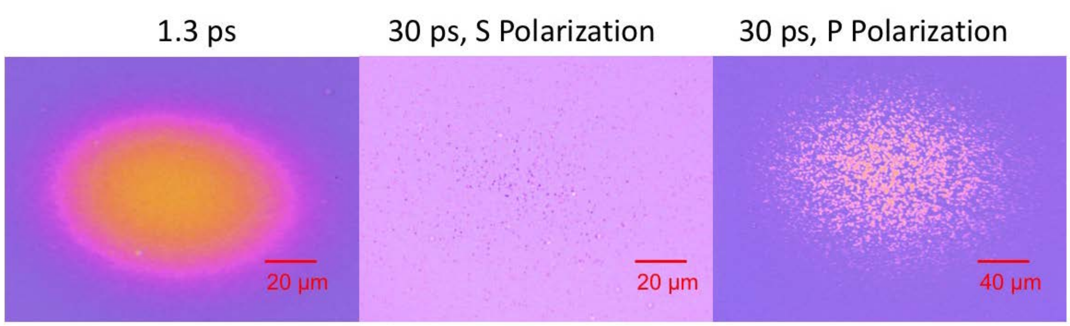

Figure 3: Damage morphology of e-beam deposited hafnia coatings as measured by confocal microscopy. Ablation craters are observed for short pulses such as the 1.3 ps pulse above. Longer pulse widths exhibit high density pits observable as small black dots for 30 ps, S polarization (9.0 J/cm^2) and isolated absorbers for 30 ps, P polarization (9.4 J/cm2).

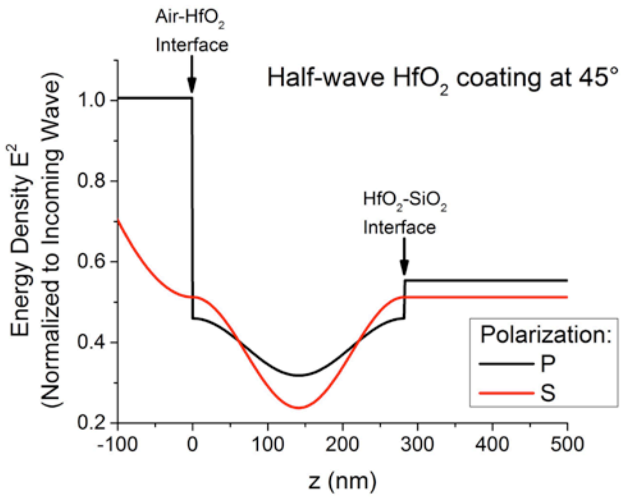

Figure 4: Electric field modeling of intensities of half-wave HfO_2 coating at 45° angle of incidence. Within the hafnia layer, the S polarization has a higher peak fluence for a given pulse energy. However, at the surface, there is a large discontinuity in the peak fluence at the surface, which allows us to distinguish between surface defects or absorbers and defects within the hafnia material. The large difference in observed damage between S and P polarization for 30 ps pulses is consistent with damage occurring due to isolated surface absorbers for hafnia.

For hafnia coatings (Figures 3 and 4), we modeled the effects of the layers on the electric field distributions for varying input polarization and incident angles. This modeling effort allowed us to distinguish between the effects of surface defects and defects within the coating materials. We found that isolated absorbers on the surface of hafnia are affected by environmental conditions, with damage suppressed by the presence of air over vacuum (we do not know if oxygen or water suppressed damage). That the chemical environment affects the progression of damage over a single 30 ps pulse indicates the complexity of the damage processes limiting the performance of these coatings.

For the second goal above, we performed multi-pulse S/1 and R/1 damage tests for hafnia and silica coatings and fused silica. S/1 tests use a pre-determined number of shots on a test site followed by a damage imaging and classification step, and R/1 tests ramp the pulse energy until damage occurs. In all cases, damage onset for short pulses (3 ps and lower) was lower for an increased number of pulses as observed elsewhere [10], indicating that either an accumulation of defects can lower the damage onset. For longer pulse widths, the multi-pulse damage tests determined at what point the damage grows and becomes important in practical applications.

For the third goal of investigating damage mechanisms and measuring key parameters related to damage, we used multiple approaches. The high-resolution optical microscopy, AFM, and SEM helped to understand the nature of the absorber distribution. For measuring fundamental parameters, the initial focus was on measuring the key parameters of avalanche ionization-related ablation (avalanche ionization, multiphoton excitation coefficients, and relaxation times). The finding that defects were dominant in the development of laser-induced damage in this regime shifted the emphasis to finding the distribution of absorbers. A key finding was that initiation of the isolated absorbers was primarily dependent of fluence, and much less dependent on pulse width (Figure 5). This indicates that the isolated absorbers are linear absorbers of energy. On the other hand, development of these isolated absorbers into growing sites is more dependent on pulse width (dotted lines in Fig. 5). For short pulses less than 3 ps, the isolated absorbers have a limited effect on damage.

Despite the emphasis on finding the distribution of absorbers in coatings, we did perform experiments that probed fundamental material limitations in bulk fused silica. We were able to obtain estimates of the parameters of avalanche ionization-related ablation in line with previous results using short-pulse results [11] and with experiments of filamentation-related damage for longer pulses in fused silica [12]. In these non-linear optical experiments, we found that filamentation-related damage

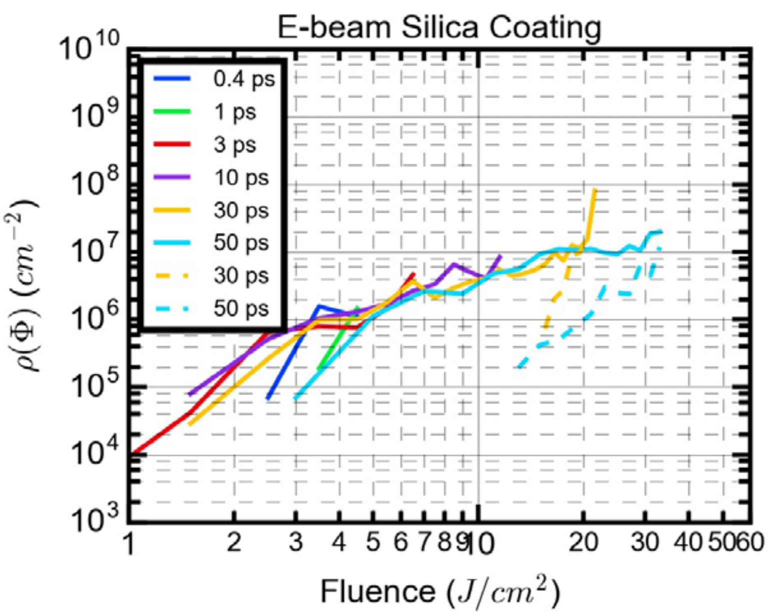

Figure 5: Density of precursors observed in e-beam silica coating as a function of pulse width and fluence. The densities observed stop when coating removal or crater formation occurs; for example, the 3 ps lines stops at about 6 J/cm^2, which is the level at which coating removal occurs. The dotted lines indicate the densities for sites that undergo subsequent damage growth. The overlap of all of these densities indicates an energy-driven rather than intensity-driven process.

occurred for pulsed 10 ps and longer, but for shorter pulses, not enough of the energy propagates in the filament to cause damage. These results help in understanding the limitations on transmitted power possible through a bulk material for ps pulses.

For the fourth goal of identifying dominant failure mechanisms and exploring mitigation strategies, we accomplished the first part through detecting the linear absorbers involved in laser damage of coatings. We performed unsuccessful annealing experiments, and ramped R/1 tests indicated some possible, marginal improvement with ramping the pulse energies. The need to reduce and eliminate the isolated absorbers indicated that a new method for depositing coatings need to be developed.

Impact on Mission

This LDRD project is having an impact on lab missions in three ways: in knowledge gained, in development of laser-damage testing facilities and procedures, and in personnel. The knowledge that a high density of defects plays an important role in laser-induced damage from 1-50 ps, especially at longer pulses, affects the path forward for reducing laser-induced damage of multi-layer optical coatings. Previously, it was found that for ns pulses, laser-induced damage thresholds were dominated by isolated, microscopic coating flaws. These flaws have been reduced through pre-initiation and planarization. We now know that there are nano-scopic, isolated absorbers at much higher densities that affect laser-induced damage threshold for 1-50 ps pulses. Damage tests at longer pulses indicate that reducing the presence of the isolated absorbers is imperative to improving the coatings. This was an important driver in the new strategic initiative (SI) LDRD grant on improving multi-layer dielectric coatings, as well as highlighting the need for the development of LLNL capabilities in ion-beam sputtering coating for improved coatings for high power lasers.

This LDRD grant was used in part to convert one postdoctoral fellow to a full-time staff scientist position, increasing the depth of expertise in high-sensitivity microscopy applied to laser-materials interactions. In addition to three existing publications [5–7] and one submitted, three additional publications related to this LDRD are in preparation.

Conclusion

The discovery that defects in the coating materials dominate the laser-induced damage performance of these materials for pulses shorter than previously believed motivates continued work to improve the materials used in multi-layer dielectric coatings. It was observed that significant improvements in damage performance require changes in the deposition processes. An SI LDRD will build on this work by aiming to improve the deposition processes used for mirror production, focusing on the longer pulse regime from 30 ps to ns. It was well known before the LDRD that at short pulses fundamental material processes lead to damage, and at long pulses the processes are defect driven. We have shown that the defects play a role at shorter pulses that previously thought: even at pulse widths down to 1 ps defects must be considered.

This work was performed under the auspices of the U.S. Department of Energy by Lawrence Livermore National Laboratory under Contract DE-AC52-07NA27344.

References

1. C. Carr, J. Bude, and P. DeMange, "Laser-supported solid-state absorption fronts in silica," Phys. Rev. B **82**, 184304 (2010).
2. B. C. Stuart, M. D. Feit, S. Herman, A. M. Rubenchik, B. W. Shore, and M. D. Perry, "Nanosecond-to-femtosecond laser-induced breakdown in dielectrics," Phys. Rev. B **53**, 1749 (1996).
3. D. A. Cross and C. W. Carr, "Analysis of 1ω bulk laser damage in KDP," Appl. Opt. **50**, D7–D11 (2011).
4. T. A. Laurence, J. D. Bude, S. Ly, N. Shen, and M. D. Feit, "Extracting the distribution of laser damage precursors on fused silica surfaces for 351 nm, 3 ns laser pulses at high fluences (20-150 J/cm< sup> 2</sup>)," Opt. Express **20**, 11561–11573 (2012).
5. D. A. Alessi, C. W. Carr, R. P. Hackel, R. A. Negres, K. Stanion, J. E. Fair, D. A. Cross, J. Nissen, R. Luthi, G. Guss, J. A. Britten, W. H. Gourdin, and C. Haefner, "Picosecond laser damage performance assessment of multilayer dielectric gratings in vacuum," Opt. Express **23**, 15532 (2015).
6. R. A. Negres, I. Bass, K. A. Stanion, G. Guss, D. A. Cross, D. A. Alessi, C. Stolz, and C. W. Carr, "Apparatus and techniques for measuring laser damage resistance of large-area, multilayer dielectric mirrors for use with high energy, picosecond lasers," in *CLEO: Science and Innovations* (Optical Society of America, 2015), p. SM3M–5.
7. R. A. Negres, C. W. Carr, T. A. Laurence, K. Stanion, G. Guss, D. A. Cross, P. J. Wegner, and C. J. Stolz, "Laser-induced damage of intrinsic and extrinsic defects by picosecond pulses on multilayer dielectric coatings for petawatt-class lasers," Opt. Eng. **56**, 011008 (2016).
8. S. Ly, N. Shen, R. A. Negres, C. W. Carr, D. A. Alessi, J. D. Bude, A. Rigatti, and T. A. Laurence, "Laser-induced modifications on the input surface of silica optics following exposure to picosecond pulses at 1053 nm," Opt. Express **Submitted**, (n.d.).
9. M. Mero, J. Liu, W. Rudolph, D. Ristau, and K. Starke, "Scaling laws of femtosecond laser pulse induced breakdown in oxide films," Phys. Rev. B **71**, (2005).
10. M. Mero, B. Clapp, J. C. Jasapara, W. Rudolph, D. Ristau, K. Starke, S. Martin, and W. Kautek, "On the damage behavior of dielectric films when illuminated with multiple femtosecond laser pulses," Opt. Eng. **44**, 051107–051107 (2005).
11. S. Tzortzakis, L. Sudrie, M. Franco, B. Prade, A. Mysyrowicz, A. Couairon, and L. Bergé, "Self-Guided Propagation of Ultrashort IR Laser Pulses in Fused Silica," Phys. Rev. Lett. **87**, (2001).
12. K. R. Manes, M. L. Spaeth, J. J. Adams, and M. W. Bowers, "Damage Mechanisms Avoided or Managed for NIF Large Optics," Fusion Sci. Technol. **69**, 146–249 (2016).