

# Dual-wavelength laser-induced damage threshold of a $\text{HfO}_2/\text{SiO}_2$ dichroic coating developed for high transmission at 527 nm and high reflection at 1054 nm

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## ABSTRACT

Dichroic coatings have been developed for high transmission at 527 nm and high reflection at 1054 nm for laser operations in the nanosecond pulse regime. The coatings consist of  $\text{HfO}_2$  and  $\text{SiO}_2$  layers deposited with e-beam evaporation, and laser-induced damage thresholds as high as  $12.5 \text{ J/cm}^2$  were measured at 532 nm with 3.5 ns pulses (22.5 degrees angle of incidence, in S-polarization). However, laser damage measurements at the single wavelength of 532 nm do not adequately characterize the laser damage resistance of these coatings, since they were designed to operate at dual wavelengths simultaneously. This became apparent after one of the coatings damaged prematurely at a lower fluence in the beam train, which inspired further investigations. To gain a more complete understanding of the laser damage resistance, results of a dual-wavelength laser damage test performed at both 532 nm and 1064 nm are presented.

Keywords: Optical Coatings,  $\text{HfO}_2$ ,  $\text{SiO}_2$ , Laser Damage, Dichroic, Dual-wavelength, High Reflection, High Transmission

## 1. INTRODUCTION

A dichroic coating [1] was developed for a beam combining application at the Z-Backlighter Laser Facility [2] at Sandia National Laboratories. The Z-Backlighter lasers are kJ-class systems that require optical coatings with high resistance to laser damage. The dichroic coating was designed for high transmission at 527 nm and high reflection at 1054 nm, at  $22.5^\circ$  in S-pol for both wavelengths, in the nanosecond pulse regime. A coating of this design does not have high resistance to laser damage, with the most vulnerable aspect being high transmission at 527 nm, in which the fluence at higher photon energy compared to 1064 nm penetrates through all the coating layers. The dichroic coating is 23 layers thick, which is necessary to satisfy the high reflection requirement at 1054 nm. This means that the higher energy, 527 nm photons can trigger damage precursors all throughout the 23-layer stack, including on the substrate.

Although the dichroic coating is designed to operate at 527 nm and 1054 nm simultaneously, prior laser damage testing addressed each wavelength separately. Using the NIF-MEL protocol and 3.5 ns duration pulses, the laser-induced damage threshold (LIDT) was  $12.5 \text{ J/cm}^2$  at 532 nm ( $22.5^\circ$  S-pol) [1]. At 1064 nm ( $22.5^\circ$  S-pol), the LIDT was  $32 \text{ J/cm}^2$  [1]. The objective of this study was to understand how the LIDT would be affected by dual wavelength testing at 532 nm and 1064 nm simultaneously.

In the literature, dual-wavelength LIDT testing has been conducted on silica substrates [3,4], antireflection coatings [5], and beam splitter coatings [6-8]. In general, dual-wavelength LIDTs were lower compared to single-wavelength results, and they depended on the temporal separation of the two pulses [4,5,8].

Lamainere, et al, [4] performed dual-wavelength LIDT tests at 355 nm and 1064 nm on amorphous silica samples (Heraeus S312). They demonstrated that damage initiation was influenced most by the 355 nm pulses, which induced preexisting damage craters to grow. Dual-wavelength illumination resulted in a higher damage density, and the onset of damage at lower fluence, compared to single-wavelength 355 nm pulses. However, the effect of the 1064 nm beam on damage growth depended on the temporal delay between the 355 nm and 1064 nm beam. If the 1064 nm pulse reached the sample before the 355 nm pulse, there was nearly no enhancement of damage growth, but if the 1064 nm pulse arrived after the 355 nm pulse, then the growth of damage was enhanced. Mrohs et al [5] conveyed similar results using a different experiment. They tested antireflection coatings at 266 nm and 532 nm and

reported higher damage densities resulting from dual-wavelength illumination, and noted that the dual-wavelength LIDT was a function of the temporal delay between the two pulses.

## 2. DICHROIC COATING DEVELOPMENT

The dichroic coating was designed using Optilayer software [9] to optimize the thicknesses of the alternating  $\text{HfO}_2$  and  $\text{SiO}_2$  layers [1, 10]. In general, the spectral requirements for the dichroic coating were challenging to meet because the index of refraction of  $\text{HfO}_2$  at 527 nm is higher compared to at 1054 nm, which means that a quarter-wave standard reflector design for 1054 nm does not equate to high transmission at the half wavelength of 527 nm. Therefore, Optilayer was used to find layer thicknesses that satisfied the competing spectral requirements at 527 nm and 1054 nm. Furthermore, the electric field intensities were tracked in Optilayer and optimized at 527 nm, however optimization resulted in some loss of spectral performance.

The first iteration of the coating design was 22 layers and was deposited using electron-beam evaporation with ion-assisted deposition (IAD) [10] in a large, custom coating system at Sandia National Laboratories [11]. This coating had a lower LIDT of  $7 \text{ J/cm}^2$  (532 nm, 22.5° S-pol, 3.5 ns, NIF-MEL protocol [10]). Since the coating design was already optimized, changes to the deposition process were implemented to increase the LIDT of the coating, with the aim of reducing defects in the coating that could act as precursors to laser damage. These changes included deposition without IAD, reducing the  $\text{HfO}_2$  deposition rate from 3 Å/s to 2 Å/s, and depositing a 100-nm  $\text{SiO}_2$  adhesion layer before the first layer of  $\text{HfO}_2$  was deposited [1]. The resulting 23-layer dichroic coating had a higher LIDT of  $12.5 \text{ J/cm}^2$  (532 nm, 22.5° S-pol, 3.5 ns, NIF-MEL damage test protocol [1]). The improved LIDT was welcome, however the spectral performance of the coating was affected because the lack of IAD decreased the refractive index of  $\text{HfO}_2$ , overall reducing the high reflection bandwidth at 1054 nm. The spectral performance (transmission) is shown below in Fig. 1. The coating hardly meets the high transmission (532 nm) and high reflection (1054 nm) requirements, and this is a tradeoff that was accepted to achieve higher LIDT.

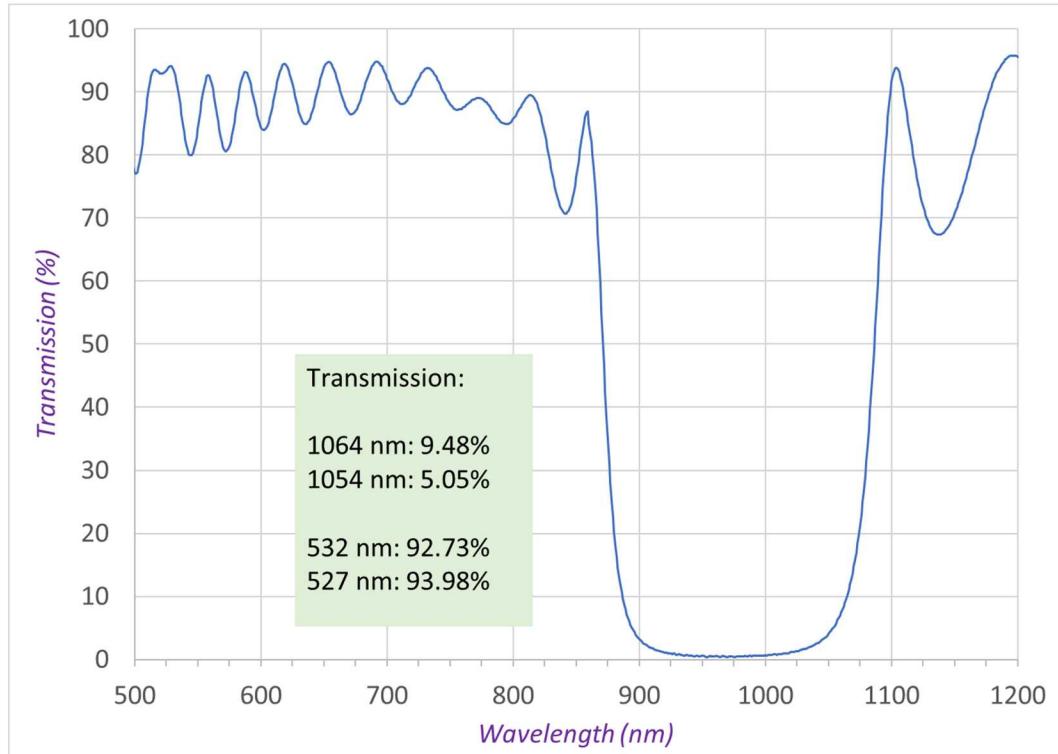


Figure 1: Spectral transmission at 22.5° in S-pol. The transmission includes the ~4% Fresnel reflection from the uncoated backside of the optic. The coating is intended for operation at 527 nm and 1054 nm, while 532 nm and 1064 nm were used for laser damage testing. The transmission scan was acquired with a Perkin-Elmer Lambda 950 spectrophotometer.

A clue into the dual-wavelength laser damage resistance of the coating can be established from electric field models. In Figs. 2 and 3, electric field intensity was modeled in Optilayer at the wavelengths of 532 nm and 1064 nm, which correspond to the wavelengths used for laser damage testing.

At 532 nm (Fig. 2), the two outermost  $\text{SiO}_2$  layers experience the highest electric field magnitudes, followed by the outermost  $\text{HfO}_2$  layer. Defects in these outer layers will therefore experience the highest fluences, but the  $\text{HfO}_2$  layer may be of particular concern due to the lower laser damage resistance of this material compared to  $\text{SiO}_2$ . Unfortunately, at 532 nm the electric field intensity remains relatively high throughout all the  $\text{SiO}_2$  layers in the coating because the coating was designed to be transmissive at this wavelength. Although the coating design had been optimized to minimize electric field intensities in the inner  $\text{HfO}_2$  layers, higher intensities still exist at many of the  $\text{HfO}_2/\text{SiO}_2$  layer interfaces, which are vulnerable to damage [7].

At 1064 nm (Fig. 3), the highest electric field magnitudes occur in the outermost  $\text{SiO}_2$  and  $\text{HfO}_2$  layers, and gradually reduce further into the coating. Therefore, defects in the outer layers of the coating are the most likely damage precursors, while defects in the inner layers are less likely to have an impact.

The combined effect of high electric field intensities at both 532 nm and 1064 nm in the outermost coating layers may result in a larger number of damage sites, since 532 nm and 1064 nm damage precursors would be affected at the same time. This could lead to lower resistance to laser damage overall compared to single-wavelength illumination. Also, due to the transmission of 532 nm fluence through the coating, damage sites may also occur in the inner coating layers or on the substrate. The 1064 nm fluence is lower in the inner coating layers, and it is unclear whether this fluence would also affect the growth of the inner 532 nm damage sites.

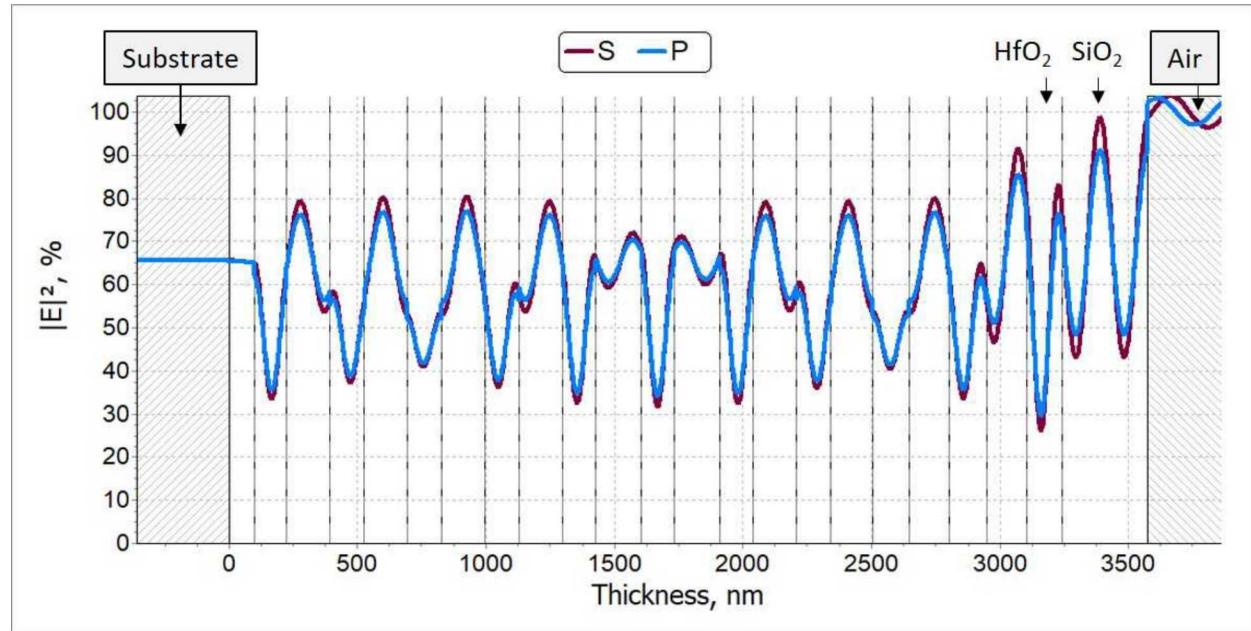


Figure 2: Electric field intensity model at 532 nm, 22.5° (high transmission). The outermost  $\text{SiO}_2$  layers experience the highest fluences, however, the electric field intensity remains relatively high throughout the coating, including at  $\text{HfO}_2/\text{SiO}_2$  interfaces. Note that the first layer of the coating deposited on the substrate is 100 nm of  $\text{SiO}_2$  and is essentially an absentee layer meant to improve adhesion of the first  $\text{HfO}_2$  layer.

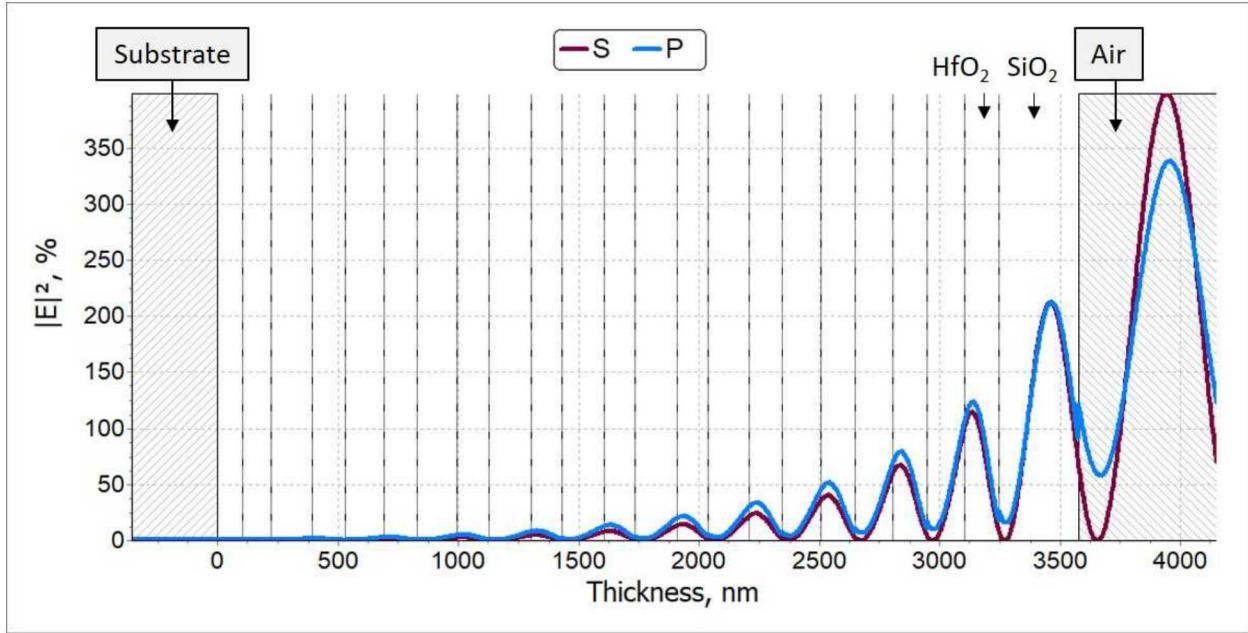


Figure 3: Electric field intensity model at 1064 nm, 22.5° (high reflection). The electric field intensity is highest only in the outermost layers due to the high reflection application at this wavelength.

### 3. EXPERIMENTAL SETUP

The dichroic coating was deposited on an optically polished fused silica substrate (50 mm diameter, 12.7 mm thick). Just prior to the coating process, the substrate was cleaned using our standard process that involves manually washing with mild Micro 90 detergent, Baikalox alumina slurry, deionized water, and clean room wipes (Texwipe TX1010) [12].

The coating was deposited at a temperature of 200°C. The SiO<sub>2</sub> layers were deposited at a rate of 7 Å/s from 1-3 mm SiO<sub>2</sub> granules. The HfO<sub>2</sub> layers were deposited from hafnium metal at 2 Å/s with O<sub>2</sub> backfill (0.9x10<sup>-4</sup> Torr total pressure in coating chamber). In addition, the coating system contained masking to maintain uniformity, and quartz crystal monitoring was used for layer thickness control. After coating, the substrate was cleaned with deionized water and Micro 90 detergent [12].

The dual-wavelength laser damage tests were conducted by Spica Technologies [13] based on the NIF-MEL protocol [14], which is a raster scan testing method consisting of 2500 sites in a 1 cm<sup>2</sup> area. Testing at single wavelength is described first to provide a basis for the setup of the dual wavelength test.

Testing at a single wavelength involves the use of single transverse mode (Gaussian), multi-longitudinal mode laser pulses of 3.5 ns duration. The repetition rate is 5 Hz and the beam diameter ( $1/e^2$ ) is 1.03 mm (at 532 nm) and 1.02 mm (at 1064 nm). In the raster scan, the laser spot overlaps itself from site to site at 90% of its peak intensity radius. The laser fluence begins at 1 J/cm<sup>2</sup> at each of the 2500 sites. After testing each site at this fluence, the fluence is increased by 1 J/cm<sup>2</sup>, and each of the 2500 sites are tested again. This process repeats until the damage threshold fluence is reached. The accuracy of the fluence measurement is +/- 3%. Also, the test occurs in a dry, 0% humidity nitrogen-filled environment.

Laser damage is identified as a melt or crater that alters the coated surface. As the laser fluence increases, some of these damage sites may grow in size. This is called propagating damage, and can lead to catastrophic damage. The damage sites that do not grow with increasing fluence are classified as non-propagating damage. Non-propagating damage is generally associated with coating defects such as nodules that eject from the coating upon illumination by a certain fluence, leaving behind a stabilized damage site below a threshold fluence [15]. On the other hand, propagating damage can be indicative of intrinsic properties of the coating that lead to damage, such as the bandgap

of the material. In the ns-regime, damage is usually dominated by the accumulation of non-propagating defects [15].

According to the NIF-MEL damage criterion, the LIDT can be established in two ways: (1) the fluence at which one or more propagating damage sites occurs, or (2) the fluence at which the number of non-propagating damage sites exceeds 25. The lowest fluence that satisfies either of those conditions is considered the LIDT. Moreover, having 25 or more non-propagating damage sites comprises 1% of the 2500 sites in the raster scan area of  $1\text{ cm}^2$ . While non-propagating damage sites may be benign and resist growth below a certain threshold, they are still flaws and can lead to undesirable effects such as scattering.

The dual-wavelength laser damage test was conducted using the NIF-MEL protocol with some modifications. The 1064 nm fluence was partially converted to 532 nm based on the following proportion: the 532 nm fluence was maintained at 80% of the 1064 nm fluence. In other words, if the 1064 nm fluence was  $10\text{ J/cm}^2$ , then the 532 nm fluence was  $8\text{ J/cm}^2$ . The fluences were orthogonally polarized, so that the 1064 nm fluence was in S-pol and the 532 nm fluence was in P-pol. The beam diameters ( $1/e^2$ ) were  $905\text{ }\mu\text{m}$  (at 1064 nm), and  $394\text{ }\mu\text{m}$  (at 532 nm). Also, the pulses overlapped in time: the 1064 nm pulse width was 3.5 ns, and the 532 nm pulse width was 2.5 ns.

#### 4. RESULTS

The dual-wavelength laser damage test results are shown in Fig. 4, along with the single-wavelength results for comparison. The dual wavelength LIDT, and the single wavelength LIDT at 532 nm, are both characterized by the occurrence of non-propagating defects. Not surprisingly, the dual-wavelength LIDT is lower than the single-wavelength LIDT when compared at the 532 nm fluence: the single wavelength LIDT is  $12.5\text{ J/cm}^2$  at 532 nm, and the dual wavelength LIDT is  $10.5\text{ J/cm}^2$  at 532 nm (with  $13.1\text{ J/cm}^2$  at 1064 nm).

The slopes of the single-wavelength (532 nm) and dual-wavelength LIDT results appear to be almost the same, which suggests that the onset of damage is still governed by the 532 nm photons, even though the 1064 nm fluence was higher. However, in the single-wavelength 1064 nm test, the first damage site occurred at  $17\text{ J/cm}^2$ , which is above the fluence range of the dual-wavelength test. Also, at the single wavelength of 1064 nm, there were fewer damage precursors in general, as demonstrated by the relatively low number of non-propagating damage sites. This is expected because there are fewer precursors that are affected by lower energy, 1064 nm photons. Also, the coating is reflective at 1064 nm and therefore most of the 1064 nm fluence does not penetrate deeply into the coating. Ultimately, in the dual-wavelength test, the LIDT was reached at a fluence that was possibly too low for 1064 nm damage precursors to matter. However, that does not mean the 1064 nm fluence had no impact at all. The fact that the dual-wavelength LIDT is lower than the single wavelength LIDT suggests that the 1064 nm fluence worsened the effect of the 532 nm fluence, causing earlier occurrence of damage that lead to lower LIDT.

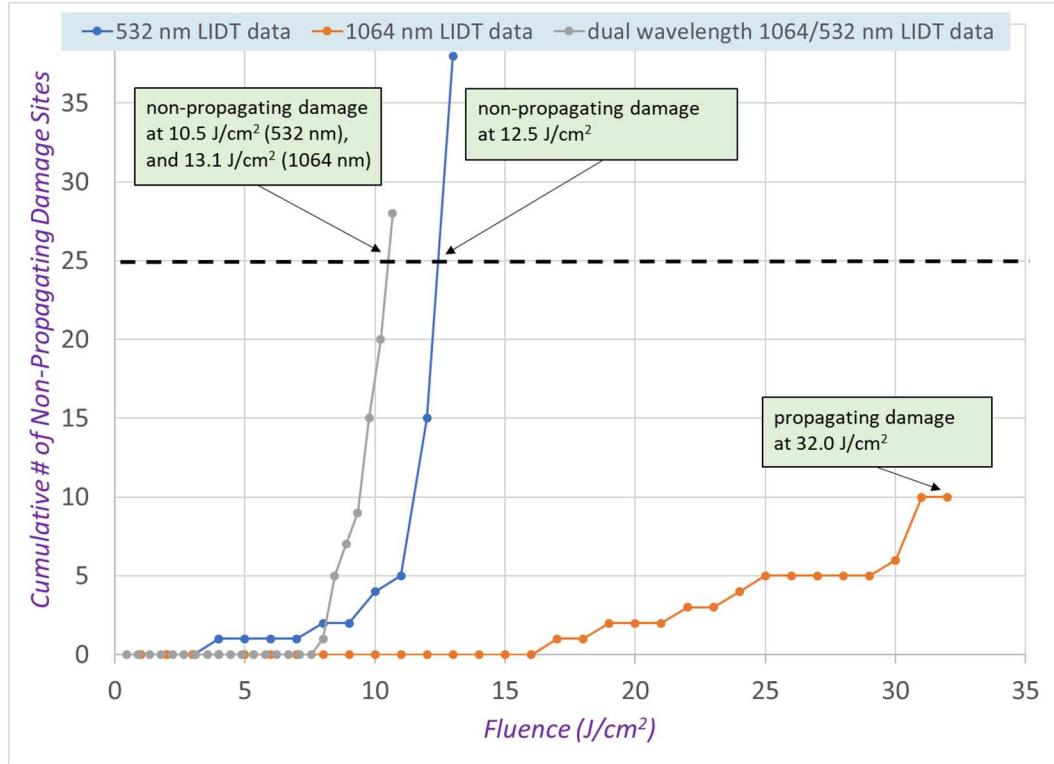


Figure 4: Laser damage testing occurred at single wavelengths [1] and dual wavelength (532 nm and 1064 nm, 22.5°). The dual-wavelength LIDT results are plotted according to the 532 nm fluence.

## 5. CONCLUSION

Dual wavelength LIDT testing was conducted on a dichroic coating designed for high transmission at 527 nm and high reflection at 1054 nm. The LIDT testing was conducted in the nanosecond pulse width regime at 532 nm and 1064 nm using a raster scan method. The dual wavelength LIDT of 10.5 J/cm<sup>2</sup> is lower than the single wavelength LIDT of 12.5 J/cm<sup>2</sup> at 532 nm. The dual wavelength LIDT was dominated by damage that was initiated by the 532 nm fluence, but the addition of the 1064 nm fluence caused damage to occur earlier, leading to lower LIDT and essentially lower tolerance to the 532 nm fluence. This indicates that damage precursors which are sensitive to 532 nm are also sensitive to 1064 nm when illuminated at both wavelengths simultaneously. Additional dual-wavelength laser damage tests using different fluence ratios for 532 nm and 1064 nm could reveal the transition points where damage is dominated by just 532 nm, 1064 nm, or both.

In future work, laser damage morphology images could reveal whether the damage occurred in the surface layers of the coating where electric field intensities are high at both 532 nm and 1064 nm, or in the internal layers where electric field intensities are high at just 532 nm. Also, investigating the effects of temporal delay between the two pulses would be useful.

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This paper describes objective technical results and analysis. Any subjective views or opinions that might be expressed in the paper do not necessarily represent the views of the U.S. Department of Energy or the United States Government.

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