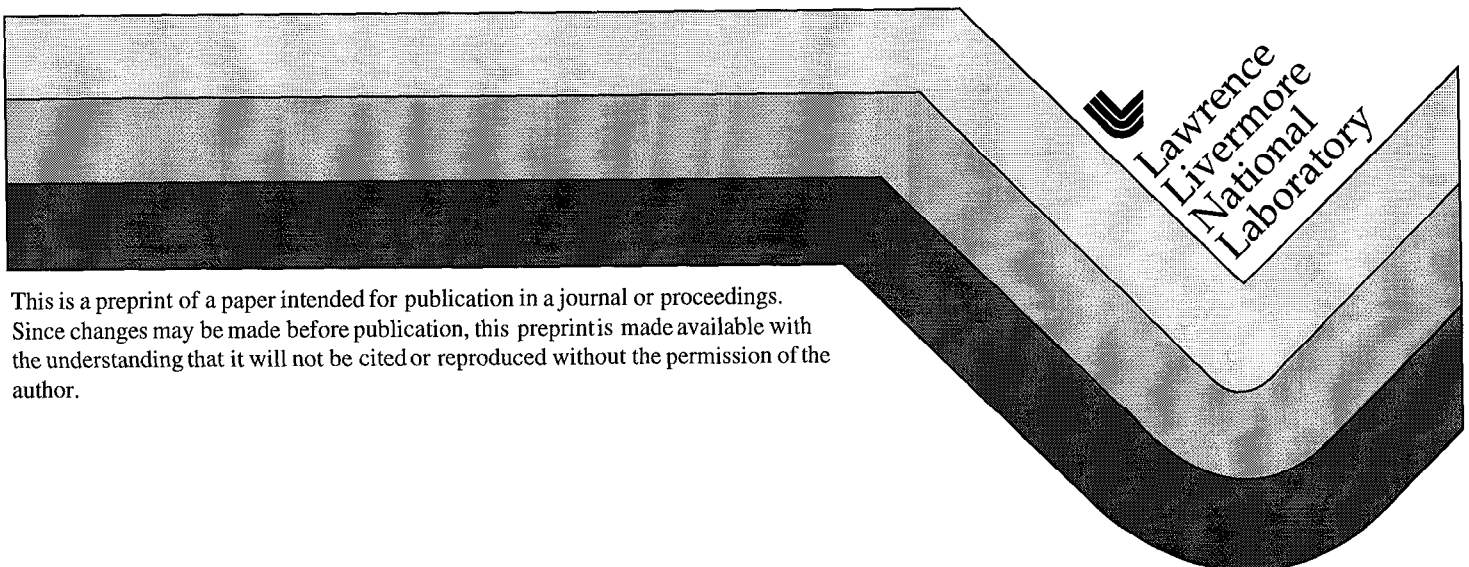# Thin Film Contamination Effects on Laser-Induced Damage of Fused Silica Surfaces at 355 nm

F. Y. Génin, A. M. Rubenchick, A. K. Burnham,
M. D. Feit, J. Yoshiyama, A. Fornier,
C. Cordillot, and D. Schirmann

July 28, 1998

DISCLAIMER

Thin film contamination effects on laser-induced damage of fused silica surfaces at 355 nm

F. Y. Génin·· , A. M. Rubenchick, A. K. Burnham, M. D. Feit,
J. Yoshiyama, A. Fornier*, C. Cordillot*, and D. Schirmann*,

University of California, Lawrence Livermore National Laboratory, Livermore, California 94550, USA.
* CEA-Centre d'Etudes de Limeil-Valenton, 94195 Villeneuve-Saint-Georges - Cédex, France.

ABSTRACT

Fused silica windows were artificially contaminated to estimate the resistance of target chamber debris shields against laser damage during NIF operation. Uniform contamination thin films (1 to 5 nm thick) were prepared by sputtering various materials (Au, Al, Cu, and B_4C). The loss of transmission of the samples was first measured. They were then tested at 355 nm in air with an 8-ns Nd:YAG laser. The damage morphologies were characterized by Nomarski optical microscopy and SEM.

Both theory and experiments showed that metal contamination for films as thin as 1 nm leads to a substantial loss of transmission. The laser damage resistance dropped very uniformly across the entire surface (e.g. 6 J/cm^2 for 5 nm of Cu). The damage morphology characterization showed that contrary to clean silica, metal coated samples did not produce pits on the surface. B_4C coated silica, on the other hand, led to a higher density of such damage pits.

A model for light absorption in the thin film was coupled with a simple heat deposition and diffusion model to perform preliminary theoretical estimates of damage thresholds. The estimates of the loss due to light absorption and reflection pointed out significant differences between metals (e.g. Al and Au). The damage threshold predictions were in qualitative agreement with experimental measurements.

Key words: UV laser damage, damage morphology, silica, contamination, cleanliness, target chamber, surface damage.

1. INTRODUCTION

The development and construction of high fluence lasers for inertial confinement fusion such as the National Ignition Facility (NIF) at Lawrence Livermore National Laboratory (LLNL) or the Laser Mégajoule (LMJ) in France is generating strong interest in the behavior of optical components under a variety of conditions that can lead to damage. It is already a challenge to prevent damage of clean optics under intense laser illumination.[1-7] For optics subject to contamination and to 355 nm and x-ray irradiation, the challenge is even higher.[8-11] In particular, the debris shields (fused silica flats) in the target chamber will be exposed to significant amounts of contamination (see Fig. 1). A preliminary analysis of the source and amount of contamination has estimated that the target will generate about 1 gram of material (e.g. Au, Cu, stainless steel, Al), the target positioner and diagnostics 0.3 gram (stainless steel and B_4C), and the beam dump 0.1 gram (stainless steel). In addition, remobilized debris from the first wall could produce 1.4 gram and the silica from damaged and ablated optics 1g. Table 1 summarizes the estimate of the weight of material and the equivalent thickness that could be deposited onto the debris shields after one NIF shot.

Finally, the refinishing and replacement rates of debris shields will have a major impact on the operating budget of NIF. Estimates have shown that the annual NIF operating cost for debris shields coating, refinishing and/or replacement could run between 4 and 20 times the capital cost for producing the 192 original debris shields.[12]

This article will first present a model describing how light is absorbed and reflected by very thin films on silica. The model will then be coupled with a simple one-dimensional calculation of heat (point source) deposition and diffusion. This thermal model will provide preliminary theoretical estimates of laser damage thresholds. The results of experimental 355-nm laser damage tests and damage morphology characterization

---

·· electronic mail: fgenin@llnl.gov

of artificially contaminated fused silica windows will then be described and compared to the theoretical calculations.
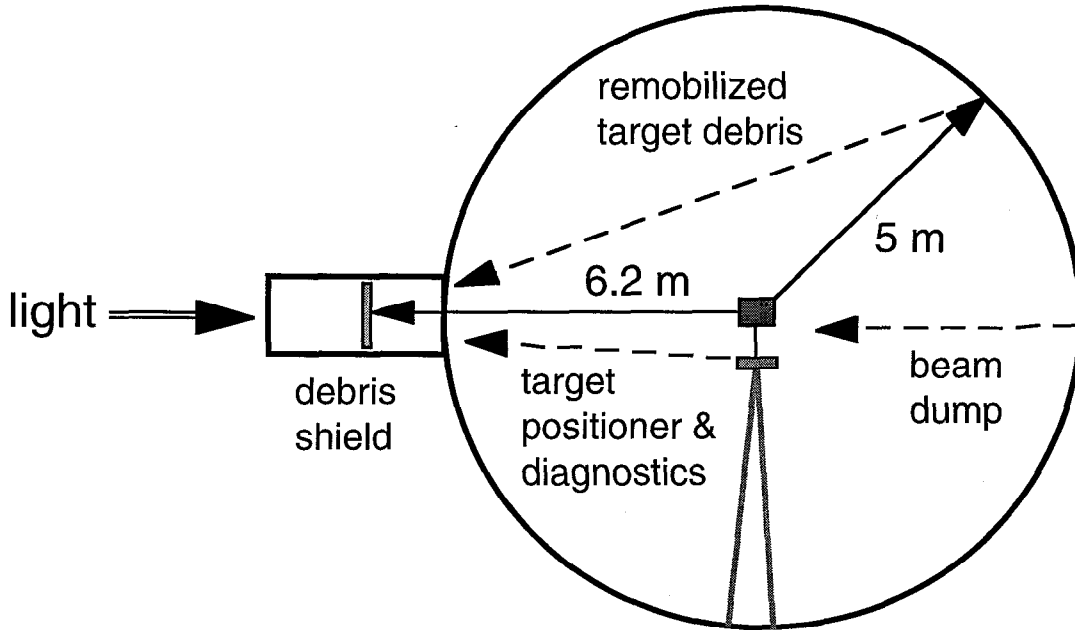

Figure 1: Schematic diagram of the target chamber showing the potential exposure of debris shields to contamination from the Holhraum, the target positioner and diagnostic, the beam dumps and remobilized debris from the first wall.

Material	Mass (μm/cm^2)	Thickness (Å)
Au	0.09	0.5
Cu	0.23	2.5
Al	0.05	1.8
SiO$_2$	0.23	10.0
B$_4$C	0.09	3.6
stainless steel	0.14	1.7

Table 1: Estimated weight of material and equivalent thickness that could be deposited onto the debris shields after each NIF shot assuming the contamination is spread evenly over 300 m^2.

In the following, the phrase "laser damage" will often be used. It is necessary to clarify its meaning within the context of this work. We usually classify damage into three categories: benign, massive and catastrophic. Benign damage describes sub-micron-size pits on the surface that are found at the onset of damaging fluences. Massive damage refers to damage that will affect the performance of the optic beyond the tolerance limits of the laser but is stable on subsequent shots at the same fluence. Finally, catastrophic damage (also named massive unstable) refers to the case where the glass begins to crack or be ablated during repetitive illumination. For fused silica irradiated at 355 nm, benign damage turns into catastrophic damage with only a very small increment in fluence (less than 1 J/cm^2). In this study, the experimental procedure was tuned to detect the onset of catastrophic damage. Further work is necessary however, to determine whether expected thin film contamination in NIF target chamber will exceed levels that would seriously deteriorate the functionality of the NIF debris shields.

2. LIGHT ABSORPTION AND REFLECTION IN VERY THIN FILMS

First, the light absorption in a very thin film is modeled. Consider a transparent silica window coated with a metal film thinner than 50 nm. The absorption and reflection of light through the metal-silica system is well described theoretically.[13] If the incident beam is normal to the surface, and the exit medium

is vacuum, the reflection coefficient, (i.e. the ratio between the amplitude of reflected to that of the incident wave) , r is given by:

$$r = \frac{\left(r_{12} + r_{23}e^{2i\phi}\right)}{\left(1 + r_{12}r_{23}e^{2i\phi}\right)},$$ (1)

where r_{ik} is the reflection coefficient interface between medium i and k, n_i is a refractive index of medium i, and indices 1,2,3 correspond to silica, metal and vacuum;

$$r_{ik} = \frac{n_i - n_k}{n_i + n_k}.$$ (2)

For silica, the refractive index n is about 1.5 and for the metal film, the refractive index n is a complex number. The phase change in the metal film Φ = khn, where k is the light wavenumber in vacuum, h-film thickness. Since khn <<1, the film transparency (i.e. the ratio of the amplitude of the transmitted to that of the incident wave), t is given by:

$$t = 4\frac{e^{i\phi}n_1n_2}{\left(n_2 + n_1\right)\left(n_2 + n_3\right)\left(1 + r_{12}r_{23}e^{2i\phi}\right)} = 6\frac{e^{i\phi}n}{\left(1 + r_{12}r_{23}e^{2i\phi}\right)\left(n+1\right)\left(n+1.5\right)}$$ (3)

and the absorption, A, is given by:

$$A = 1 - |r|^2 - \frac{n_3}{n_1}|t|^2 = 1 - |r|^2 - \frac{1}{1.5}|t|^2$$ (4)

The complex refractive index n in the UV is not often available. Furthermore, substantial variations are found in the published literature. These can be expected from differences with bulk data and because the refractive index can be sensitive to the film preparation method. The data reported in Table 2 and used to compare model with experiments should therefore be treated only as indicative.

Material	Refractive index at 355 nm
Au	1.74+1.85i
Al	0.38+4.3i
Cu	1.31+1.87i
Ag	0.2+1.44i

Table 2: Complex refractive index at 355 nm for gold, aluminum, copper,and silver.[14]

Figure 2 shows the reflection and absorption coefficients as a function of thickness at 355 nm. The calculations are performed for four metals: gold, aluminum, copper, and silver. As anticipated from the values of n in Table 2, the calculations show that aluminum absorbs and reflects very differently from gold, copper and silver, which behave in a similar fashion. It should be emphasized that if oxidation of the metal occurs, the results can be significantly affected. For example, aluminum quickly forms a transparent surface oxide with a thickness of about 3.5 nm, and a plot of transmittance versus thickness extrapolates to zero at that thickness.
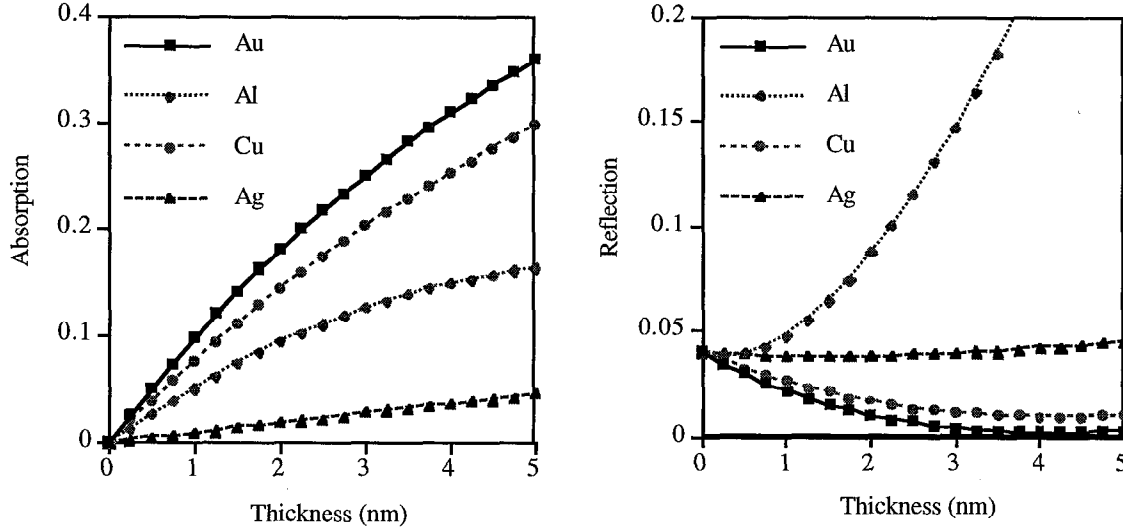
a) Absorption   b) Reflection

Figure 2: a) Reflection and b) absorption coefficients as a function of thickness for Au, Al, Cu, and Ag at 355 nm. Note that the scales are different for these two graphs.

3. ENERGY DEPOSITION AND HEAT DIFFUSION INTO SILICA

Several mechanisms can initiate damage in silica. Film heating can generate thermo-elastic stresses and crack the glass. This effect may be intensified by producing stress gradients if the film is not uniform. The absorbing metal film first heats up the glass. Above a critical temperature (e.g. 1500°C), silica begins to absorb the laser radiation more effectively. The absorption zone propagates upstream with the laser-supported detonation wave. The exploding film can then generate high pressures at the glass-film interface. If the film is not homogeneous, the transversal pressure gradient can crack the surface. Also, UV radiation from the plasma flash may produce F-centers in the glass which can later absorb the energy during subsequent pulses. Noticeable reflection from the film can produce local field intensification via interference with the incident wave stimulating damage initiation. Glass damage can take place if the glass temperature exceeds a critical value at which it is no longer transparent or melting could take place. This critical temperature is assumed to be between 1000°C and 2000°C.

The energy absorbed in the film is the product of the fluence of laser pulse, F, and the absorption coefficient, A. This energy is distributed in a layer thickness, l, written as:

$$l = 2\sqrt{\chi\tau}.$$

(5)

where χ is the thermal diffusivity and τ is the pulse length. Since the pulse is very short, this layer is very thin and the temperature in the film is very uniform. The heat source can therefore be approximated as a point source. The energy to evaporate the thin metal layer is small compared to that transferred to the glass. The temperature of the glass is therefore given by:

$$T = \frac{AF}{\rho c\sqrt{\chi\tau}} = \frac{AF}{\sqrt{\tau}}10^5\,K,$$

(6)

where the fluence is expressed in J/cm^2 and the pulse duration in nanoseconds. Equation (6) can only give an approximate estimate of the glass temperature. However, an absorption level of only 1% with a 3-ns, 5 J/cm^2 pulse can heat the surface of a silica layer to 2,900K (above the critical temperature of enhanced absorption and above the glass transition temperature).

4

4. EXPERIMENTAL RESULTS

4.1 Experimental procedure:

Laser damage tests were carried out at 355 nm using an 8.4-ns Nd:YAG laser in S-polarization at 5° and 20° incidence angle using a 1/1 (single shot) and R/1 (ramping test) procedure. The laser was focused to provide a Gaussian beam with a diameter of 1.2 mm at $1/e^2$ peak intensity. Damage was detected by monitoring the scattering of a HeNe laser focused on the output surface of the tested site; a site was declared damaged when light scattering reached a given threshold. In this study, the damage detected by the HeNe was catastrophic. The R/1 laser damage maps were obtained to measure the average LIDT, the standard deviation, the lowest and higher LIDT. While the average damage threshold is a good measure of the overall quality of an optic, the standard deviation provides information on the uniformity of the material and process.[15, 16] Finally, the lowest threshold to failure is an important engineering parameter for the laser since it will determine the damage threshold and the functionality of large-area optics. Damage for the 1/1 tests were characterized by Nomarski optical microscopy and SEM.

A set of Corning UV grade fused silica blanks 11 mm thick and 50 mm in diameter were polished using a medium and a high LIDT polishing process. The samples were then cleaned at LLNL using the NIF standard cleaning procedure. A set of samples was contaminated by sputtering material (Al, Au, Cu, and B_4C) onto the surface. The tests were performed with the contamination film on the output surface.

4.2 Damage threshold measurements:

The R/1 damage threshold measurements were performed on two polished bare fused silica windows and on fused silica contaminated with 5 nm of Cu. Twenty sites were measured for each sample. The average LIDT for the two polished silica samples were 20.1 and 27.6 J/cm^2, respectively. The contaminated samples damaged at 10.5 and 11.0 J/cm^2, respectively. The typical standard deviation for clean silica and for the contaminated silica was about 3.2 J/cm^2, and 0.4 J/cm^2.

For all other samples, the presence of a contamination layer also led to very uniform damage behavior. Low fluences shots on a contaminated surface systematically ignited plasmas. Although it was not quantified, all tested samples (even with the 1 nm deposits) showed a substantial degradation in damage resistance.

4.3 Damage morphology characterization

The damage morphology characterization of the samples showed that the surface of a clean silica after irradiation is covered with ellipsoidal micro-pits.[17] The sample with a higher LIDT had a much lower micro-pit (i.e. damage precursor) density (see Fig. 3).[16]

a) medium damage threshold polish   b) higher damage threshold polish

Figure 3: Nomarski optical micrographs of output damage on a clean surface after a single pulse in "P" polarization in air at 355 nm at about 30 J/cm^2; a) medium damage threshold surface with a high density of damage initiators and b) higher damage threshold surface with fewer cracks.

The bare surface, the Cu and B_4C film contamination samples led to different damage morphologies (see Fig. 4). At the onset of damage, a medium density of micron-size pits was found after irradiating the clean silica surface. Micro-scratches were not usually revealed. A higher pit density was detected for the B_4C and fine scratches were outlined. The surface damage pits were smaller than for clean surfaces after damage. Finally, no pits were found on metal contaminated surfaces (Cu, Au, and Al); however, scratches were also outlined. The areas damaged at higher fluence on metal contaminated surface showed strong evidence of surface melting.
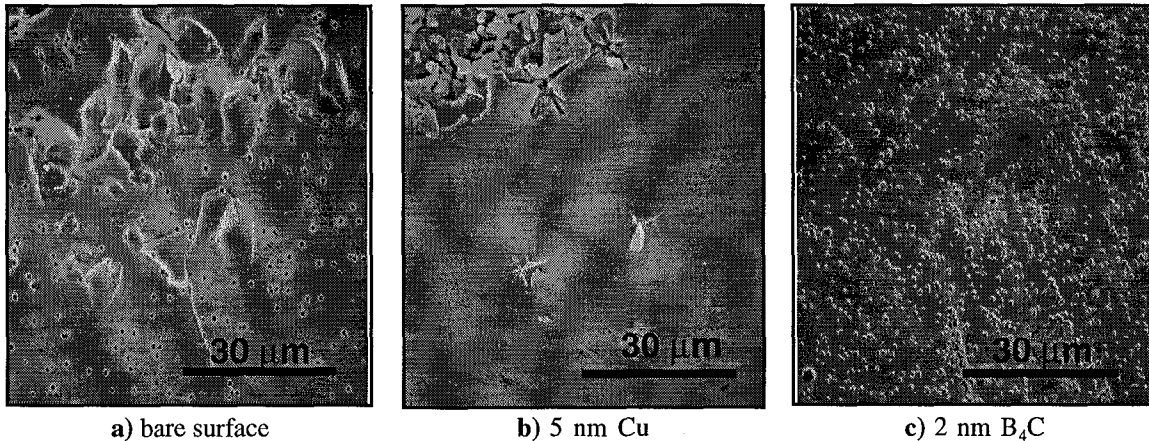

| a) bare surface | b) 5 nm Cu | c) 2 nm B_4C |

Figure 4: SEM micrographs of output surface damage after a single 8.4-ns pulse in "P" polarization in air at 355 nm: a) clean surface at 20 J/cm^2, b) surface coated with 5 nm of Cu at 20 J/cm^2, and c) surface coated with 5 nm of B_4C at 25 J/cm^2.

5. CONCLUSION

Fused silica windows were artificially contaminated with various materials (Au, Al, Cu, and B_4C). A substantial loss of transmission (greater than 1%) was measured for metal contamination films as thin as 1 nm. The damage tests at 355 nm in air with an 8-ns Nd:YAG laser showed that the damage resistance of contaminated fused silica drops very uniformly across the entire surface.

Theoretical calculations of absorption and reflection due to the presence of contamination confirmed that very thin layers could greatly reduce transmission and initiate damage at the surface. For example, a gold film, 1 nm thick, can absorb as much as 10% of the incident energy. Substantial differences between metals (e.g. gold and aluminum) were predicted.

A simple heat absorption and diffusion model showed that a 3-ns, 5 J/cm^2 laser pulse could initiate damage if contamination was to absorb only one percent of the energy. These predictions were in qualitative agreement with the experimental measurements.

Finally, the damage morphology characterization showed that contrary to clean fused silica, metal coated samples did not produce micro-pits on the surface. B_4C coated silica, on the other hand, led to a higher density of such damage pits.

To summarize, these results clearly confirm that at high fluence, trace level contamination is potentially very harmful to the integrity of transmissive fused silica optics. Further evaluation of contamination-initiated laser damage is necessary to better predict the lifetime of NIF final optics assembly.

6. ACKNOWLEDGMENTS

7. REFERENCES

1. N. L. Boling, G. Dubé, M. D. Crisp, "Laser surface damage studies on several glasses", in Laser Induced Damage in Optical Materials, *NBS-SP* **387**, 69 (1973).

2. N. L. Boling, J. A. Ringlien, and G. Dubé, "Q-switched laser induced surface damage at 1 μm", in Laser Induced Damage in Optical Materials, *NBS-SP* **414**, 119 (1974).

3. R. A. House, J. R. Bettis, and A. H. Guenther, "Subsurface structure and laser damage threshold", *IEEE J. of Quant. Electronics* **QE-13**, 363 (1977).

4. H. Vora, R. H. Anderson, and R. J. Stokes, "Surface finishing using soft abrasives", in Laser Induced Damage in Optical Materials, *NBS-SP* **638**, 262 (1981).

5. H. Vora, W. H. Lowdermilk, and J. E. Swain, "Effect of surface finishing on the damage threshold of fused silica at 1.06 μm", in Laser Induced Damage in Optical Materials, *NBS-SP* **669**, 146 (1982).

6. Y. Namba, "Float polishing and defects of fused silica", in Laser Induced Damage in Optical Materials, *NBS-SP* **669**, 138 (1982).

7. Y. Namba, M. Saeki, M. Shiokawa, K. Yoshida, et al., "Ultraviolet-laser-damage threshold of optical glasses finished by various methods", in Summaries of Papers Presented at the Conference on Lasers and Electro-Optics, *IEEE Cat. No. 95CH35800*, 410 (1995).

8. F. Y. Génin, K. Michlitsch, J. Furr, M. R. Kozlowski, and P. Krulevitch, "Laser-induced damage of fused silica at 355 and 1064 nm initiated by aluminum contamination particles on the surface", in Laser-induced damage in optical materials, *SPIE* Vol. **2966**, 126 (1996).

9. M. D. Feit, A. M. Rubenchik, D. R. Faux, R. A. Riddle, A. B. Shapiro, D. C. Eder, B. M. Penetrante, D. Milam, F. Y. Génin, and M. R. Kozlowski, "Modeling of laser damage initiated by surface contamination", in Laser-induced damage in optical materials, *SPIE* Vol. **2966**, 417 (1996).

10. J. Dijon, P. Garrec, N. Kaiser, and U.B. Schallenberg, "Influence of substrate cleaning on LIDT of 355 nm HR coatings", in Laser-induced damage in optical materials, *SPIE* Vol. **2966**, 178 (1996).

11. D. J. Gallant, M. Law, and B. Pond, "Effect of cleaning on the optical absorption of calcium fluoride and fused silica at 351 nm", in Laser-induced damage in optical materials, *NIST SP* **752**, 159 (1986).

12. P. Whitman, private communication.

13. M. Born, E. Wolf, in *Principles of optics*, Permagmon Press, Oxford, 1959.

14. E. D. Palik, Handbook of optical constant of solids, Academic Press, Orlando, 1985.

15. J. Hue, F. Y. Génin, S. M. Maricle, and M. R. Kozlowski, "Toward predicting the laser damage threshold of large-area optics", in Laser-induced damage in optical materials, *SPIE* Vol. **2966**, 451 (1996).

16. F. Y. Génin, L. M. Sheehan, J. Yoshiyama, J. Dijon, and P. Garrec, "Statistical study of UV-laser-induced failure of fused silica", *Laser-induced damage in optical materials, SPIE* Vol. **3244**, 155 (1997).

17. J. Yoshiyama, F. Y. Génin, A. Salleo, I. M. Thomas, M. R. Kozlowski, L. M. Sheehan, I. D. Hutcheon and D. W. Camp, "Effects of polishing, etching, cleaving and water leaching on the UV laser damage of fused silica", *Laser-induced damage in optical materials, SPIE* Vol. **3244**, 331 (1997).