CONF-8009119--1

LA-UR-80-3429

MASTER

TITLE: MULTIPLE-SHOT LASER DAMAGE THRESHOLDS OF ULTRAVIOLET REFLECTORS AT 248 AND 308 NANOMETERS

AUTHOR(S): Stephen R. Foltyn
Brian E. Newnam

SUBMITTED TO: Proceedings of the 12th Annual Symposium on Optical Materials for High Power Lasers
Boulder, CO - September 30-October 1, 1980

University of California

LOS ALAMOS SCIENTIFIC LABORATORY
Post Office Box 1663 Los Alamos, New Mexico 87545
An Affirmative Action/Equal Opportunity Employer

Form No. 836 R3
St. No. 2629

UNITED STATES
DEPARTMENT OF ENERGY

MULTIPLE-SHOT LASER DAMAGE THRESHOLDS
OF ULTRAVIOLET REFLECTORS AT 248 AND 308 NANOMETERS

Stephen R. Foltyn and Brian E. Newnam
Los Alamos Scientific Laboratory
Los Alamos, New Mexico 87545

Multiple-shot damage thresholds of dielectric reflectors have been measured at 248 and 308 nm. Standard irradiation conditions were a 10-ns pulsewidth, 0.6-mm spot diameter and 35-Hz pulse repetition frequency. The reflectors, from various sources, were composed of oxide and fluoride films.

Although damage was generally initiated at visible film defects, there was no correlation between damage susceptibility and the appearance of these defects. At levels near threshold, damage was most often observed as an increase in white-light scatter of a site with no growth upon continued irradiation; at higher levels, the damage site grew with successive shots.

Test sites were subjected to at least 10^3 shots and some sites received as many as 2.5×10^4 shots; however, with only one exception damage was found to occur within the first few shots or not at all.

Reflectors at 248 nm typically had damage thresholds in the 1.0-1.8 J/cm^2 range with two samples exhibiting unexpectedly high thresholds of 2.8 and 3.0 J/cm^2. In some cases, a subthreshold pre-irradiation treatment resulted in a 20-25% enhancement in damage resistance.

Key words: Excimer lasers; fluoride coatings; laser damage; multiple-shot damage; oxide coatings; pre-irradiation; thin films; ultraviolet reflectors

1. Introduction

An area of interest to Inertial Confinement Fusion and Molecular Laser Isotope Separation programs is the development of damage-resistant optical coatings. In order to address the problem of improving reflectors for wavelengths applicable to these programs, we have recently concluded a series of multiple-shot damage threshold measurements on multilayer coatings designed for 248 and 308 nm. The purpose of this initial effort was to evaluate some candidate materials and to collect useful information for optimizing coatings made from these materials.

2. Experimental details

2.1. Test specimens

As presented in table 1, the coatings - all quarter-wave dielectric stacks - were provided by both commercial vendors and research organizations. Various oxide and fluoride materials were employed. To preclude any possible variations in the coating damage thresholds due to substrate material and roughness, all but a few coatings were deposited on 38 mm × 5 mm Suprasil 2 substrates which had been batch polished by Laser Optics, Inc. to a roughness of ~ 12 A RMS.

Table 1. Description of test specimens.

<u>Coating suppliers</u>

Design Optics
Lambda/Airtron
National Research Council, Canada
Northrop
Optical Coating Laboratories, Inc.

<u>Coating materials</u>

PbF_2/Na_3AlF_6
HfO_2/SiO_2
Al_2O_3/NaF
Sc_2O_3/MgF
ThF_4/Na_3AlF_6

2.2. Test facility

Our experiment, shown schematically in figure 1, utilized a pinhole/detector combination to directly measure the peak fluence (J/cm^2) in the incident beam. In order to account for changes in pulse energy or focusability during a test, a ratio technique was used whereby part of the input beam was focused through a pinhole to continuously monitor peak fluence A. During calibration before testing the samples, a similar arrangement at B measured the test fluence. Thus, the ratio B/A was determined. For the threshold tests, pinhole/detector B was replaced by the test sample and the test fluence was computed, while running, by using fluence A, the calibration ratio B/A and the filter transmission. The laser-induced damage to the coatings was observed visually with the aid of a 25 to 100X microscope and bright white-light illumination.

It was fortunate that a commercial laser proved useful for these tests in that the device has provided, thus far, turnkey operation for tens of millions of shots. Table 2 lists details of the laser as well as applicable test conditions. It should be noted that, while the laser is capable of operating at up to 150 Hz, detector limitations dictated an upper limit of 35 Hz. Tests were conducted at this limit in order to accumulate many shots on a test site in a short time.

Table 2. Test conditions

Laser	Lumonics 861 Multigas Excimer System operating on KrF (248 nm) or XeCl (308 nm) at pressure, voltage, and mixture specified by manufacturer
Single-pulse energy	250 mJ (248 nm); 80 mJ (308 nm)
Pulse repetition frequency	35 Hz all tests
Pulse length	10-12 ns
Mean spot diameter	0.62 mm (248 nm); 0.66 mm (308 nm)

2.3. Beam characterization

In reporting any damage threshold measurements, proper characterization of the beam at the sample location is a necessity. Figures 2 and 3 are spatial profiles of the focused beam obtained by pinhole scans in the plane normally occupied by the coating under test. The mean value of the spot diameter (I_0/e^2 level) determined from these profiles was 0.62 mm. We also monitored the temporal pulse, as shown in figure 4, which had a nominal width of 12 ns FWHM.

2.4. Characterization of damage and damage thresholds

At each test fluence 10 sites were irradiated. These sites were distributed over the entire surface of the reflector. At levels near threshold, damage was most often observed as the enlargement of an already present defect. Some sites grew with continued irradiation. At levels appreciably over threshold, a "burn pattern" of the beam profile occurred accompanied by rapid growth and flaking on successive shots. The damage threshold in these tests was defined as the highest fluence at which 10 of 10 sites survived 1000 shots without damage. In addition, we defined an "upper limit" which is the highest fluence at which at least 1 of 10 sites survived 1000 shots without damage. Of special note is that in all cases but one, damage occurred within the first five shots or it did not occur at all. Discussion of this one exception will follow later in this report.

3. Results

Typical data - in this case for a 248-nm reflector composed of the Sc_2O_3/MgF_2 quarter-wave layer combination - are presented in figure 5. According to our definitions, the damage threshold is 1.7 J/cm^2 and the upper limit is 2.6 J/cm^2. This type of threshold distribution provides even more information than the thresholds themselves. The result that 2 of 10 sites survived at 2.6 J/cm^2 is a measure of the uniformity of the coating's damage resistance and the potential performance of this design. The slope of the fitted line in figure 5 then indicates the degree to which this reflector is approaching its potential.

There was a wide variation in the slopes observed during these tests. Figure 6 is an example of two 248-nm reflectors of Al_2O_3/NaF layers produced in the same coating deposition. Of the six reflectors produced in this series, Run 1, No. 1 (solid line) has the highest threshold. It also had the steepest slope. Our overall observation has been that of every set of comparable reflectors, the highest damage threshold belonged to the reflector with the steepest slope.

Another type of nonuniformity, of macroscopic dimensions, was observed in one case. Figure 7 presents the data obtained for a single 248-nm reflector of ThF_4/Na_3AlF_6 design. The performance was quite good as indicated by the dotted line (a companion reflector has a steeper slope and a damage threshold of 3.0 J/cm^2), but one portion of this reflector, comprising about one-third of the total surface area, exhibited weaker behavior. The cause is unknown and no other samples exhibited similar properties. However, this example demonstrates the importance of sampling sites over the entire surface.

The results of all tests in this series are compiled in table 4.

Table 4. Results

248 nm Reflectors

Materials	Design	R	Threshold (J/cm^2)10/10	Upper Limit (J/cm^2)1/10	Comments
PbF_2/Na_3AlF_6	$S(LH)^{10}$	0.740	0.03	-	fogged
HfO_2/SiO_2	$S(HL)^8H$	0.978	0.8	1.0	
HfO_2/SiO_2	$S(HL)^8H$, MgF_2 Overcoat	0.984	1.0	1.4	
Al_2O_3/NaF		0.452	0.3	0.6	peak-shifted, fogged, sub. unk.
Al_2O_3/NaF		0.105	0.4	1.1	badly fogged, sub. unk.
Al_2O_3/NaF	$S(HL)^{20}H$	0.959	1.0	2.1	run 1, Si substrate
Al_2O_3/NaF	$S(HL)^{20}H$	0.961	1.1	2.1	run 1, Si substrate
Al_2O_3/NaF	$S(HL)^{20}H$	0.972	1.2	2.6	run 2
Al_2O_3/NaF	$S(HL)^{20}H$	0.966	1.3	2.3	run 2
Al_2O_3/NaF	$S(HL)^{20}H$	0.966	1.3	2.4	run 1
Al_2O_3/NaF	$S(HL)^{20}H$	0.919	1.7	2.2	run 1, slightly fogged
Sc_2O_3/MgF_2	$S(LH)^{13}$	0.976	1.7	2.6	
Sc_2O_3/MgF_2(45°)	$S(LH)^{13}$	0.966	1.8	2.5	tested at 45°
Sc_2O_3/MgF_2(45°)	$S(LH)^{13}$	0.836	1.0	1.3	tested at 0°
ThF_4/Na_3AlF_6	$S(HL)^{17}H$	0.961	2.8	3.9	run 1
$ThF_4/Na_3/AlF_6$	$S(HL)^{17}H$	0.949	3.0	3.4	run 2, different dep. cond.
			308 nm Reflectors		
ThF_4/Na_3AlF_6	$S(HL)^{19}H$	0.833	< 0.4	-	peak-shifted, delayed damage
HfO_2/SiO_2	$S(HL)^8H$	0.976	1.6	2.4	run 1
HfO_2/SiO_2	$S(HL)^8H$	0.976	2.2	3.6	run 2, different dep. cond.
HfO_2/SiO_2	$S(HL)^8HL^2$	0.961	2.0	2.8	

4. Discussion of results

Salient points about the reflector designs are:

1. PbF_2/Na_3AlF_6 Very weak, probably no contender.

2. HfO_2/SiO_2 Relatively weak, but possessed high reflectance. Provided the only direct comparison of wavelength scaling in these tests in that fluence threshold was $\propto \lambda^4$, as previously observed by Newnam and Gill [1].

3. Al_2O_3/NaF Design motivated by the high threshold measured by Newnam and Gill in 1978 [1] with [illegible]-ns pulses at 266 nm. Ignoring the fogged specimens of unknown design, the rest are in the 1.0-1.7 J/cm^2 range (comparable to the previous 266-nm values when scaled per $t^{1/2}$ and λ^4). Two coatings deposited on very smooth (3-4 A rms) Si substrates appeared slightly weaker, possibly due to coating stresses induced by higher thermal expansion of Si as compared to SiO_2.

4. Sc_2O_3/MgF_2 High thresholds. Resistance of 45° reflector tested at 0° was lower by half indicating the effect of large deviations from correct coating thicknesses.

5. ThF_4/Na_3AlF_6 Highest thresholds, even for the weak area of one reflector discussed above. Hygroscopic nature could limit its application unless well protected.

Since the thresholds for this last reflector design were very high at 248 nm, we must address the poor performance of this same design at 308 nm. The comments in table 4 provide a clue. The reflectance peak of this reflector was shifted so that R at 308 nm was only 83% and T nearly 10%. This was caused by a slight error in coating thicknesses during the evaporation procedure. The result is that a significantly larger amount of laser energy at 308 nm penetrated into the inner layers than would have occurred for a properly tuned reflector design. This also would explain the observation illustrated in figure 8 of delayed damage after the start of irradiation. At nearly any fluence level damage did occur, but only after a certain number of shots. The onset of damage was heralded by a color change, the result of interference caused by the probable separation of the coating layers. Continued irradiation led to catastrophic flaking of the coating. It has been reported by several researchers [2,3,4] that a considerable amount of water exists between layers and at the film - substrate interface of the ThF_4 films. This fact and our observations lead us to speculate that laser heating of this interlayer water caused enough stress to rupture the coatings. In the 248-nm reflectors of these materials the coating thicknesses were correctly tuned as quarter waves resulting in high reflectance. In this case, laser energy density diminishes rapidly with depth into the coating stack, and the water in the outer layer simply outgasses without problematic stress.

5. Pre-irradiation conditioning

One final topic was addressed briefly in these studies: the effect of preconditioning on the damage threshold of a particular test site. By irradiating a site with 1000 shots at threshold before actually ascertaining the damage level, an improvement was observed in most of the reflector designs tested. The results shown in figure 9 for the best Al_2O_3/NaF reflector were typical in that a 20-25% threshold increase was obtained and the slope remained the same. One example of each coating material combination was preconditioned, and the results for these are listed in table 5. All materials except ThF_4/Na_3AlF_6 exhibited improved thresholds. However, the 248-nm HfO_2/SiO_2 reflector showed no effect while the greatest improvement was observed in a 308-nm reflector of the same design.

Table 5. Effect of preconditioning.

	Material	Effect
248 nm reflectors		
	Al_2O_3/NaF	threshold increased 24% (1.7 J/cm^2 to 2.1 J/cm^2)
	Sc_2O_3/MgF_2	threshold increased 22% (1.8 J/cm^2 to 2.2 J/cm^2)
	$HfO_2/SiO_2, MgF_2$	threshold increased 20% (1.0 J/cm^2 to 1.2 J/cm^2)
	HfO_2/SiO_2	no effect
	ThF_4/Na_3AlF_6	no effect
308 nm reflectors		
	HfO_2/SiO_2	threshold increased 27% (2.2 J/cm^2 to 2.8 J/cm^2)
	ThF_4/Na_3AlF_6	no effect

We speculate that the preconditioning causes evolution of adsorbed contaminants from the reflector surface thereby reducing the absorption of laser energy. However, we have not yet determined if the resultant improvement in the damage resistance is permanent or temporary. More investigation of this effect is clearly motivated.

6. Conclusions

In our tests of 248-nm and 308-nm dielectric reflectors we have observed damage thresholds of 1.0-3.0 J/cm^2. In addition, we have observed wide variations in site-to-site damage thresholds, supportive evidence for λ^4 scaling and the adverse effect of incorrect layer thicknesses. We have shown that in some materials a subthreshold pre-irradiation treatment results in a 20-25% improvement in damage resistance.

References

[1]. Newnam, B. E., and Gill, D. H., Ultraviolet damage resistance of laser coatings, NBS Spec. Pub. 541, 190 (1978).

[2]. Donovan, T.M., and Temple, P. A., The relative importance of interface and volume absorption by water in evaporated films, NBS Spec. Pub. 568, 237 (1979).

[3]. Humpherys, T. W.; Lusk, R. L.; and Jungling, K. C., Surface microanalysis techniques for characterization of thin films, NBS Spec. Pub. 568, 257 (1979).

[4]. Anderson, W. J., Hanson, W. N., Characterization of small absorptions in optical coatings, poster presentation at Twelfth Annual Symposium on Optical Materials for High Power Lasers, Boulder, Colorado (Sept. 30-Oct. 1980).

Figure Captions

Figure 1. Schematic of ultraviolet laser damage experiment.

Figure 2. Spatial profile of focused 248-nm laser beam (horizontal).

Figure 3. Spatial profile of focused 248-nm laser beam (vertical).

Figure 4. Laser temporal pulsewidth at 248 nm. FWHM is 12 ns.

Figure 5. Damage versus laser fluence at 248 nm.

Figure 6. Damage versus laser fluence at 248 nm.

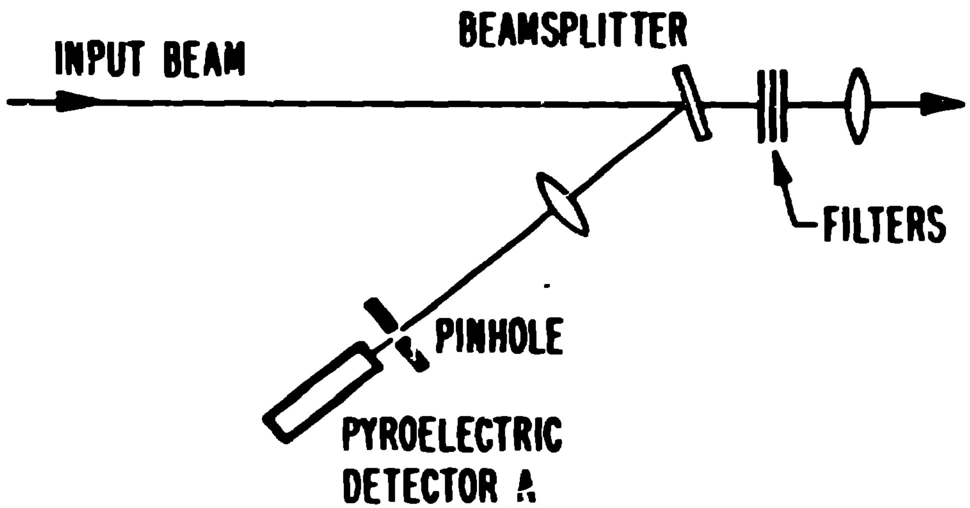
Figure 7. Damage versus laser fluence at 248 nm.

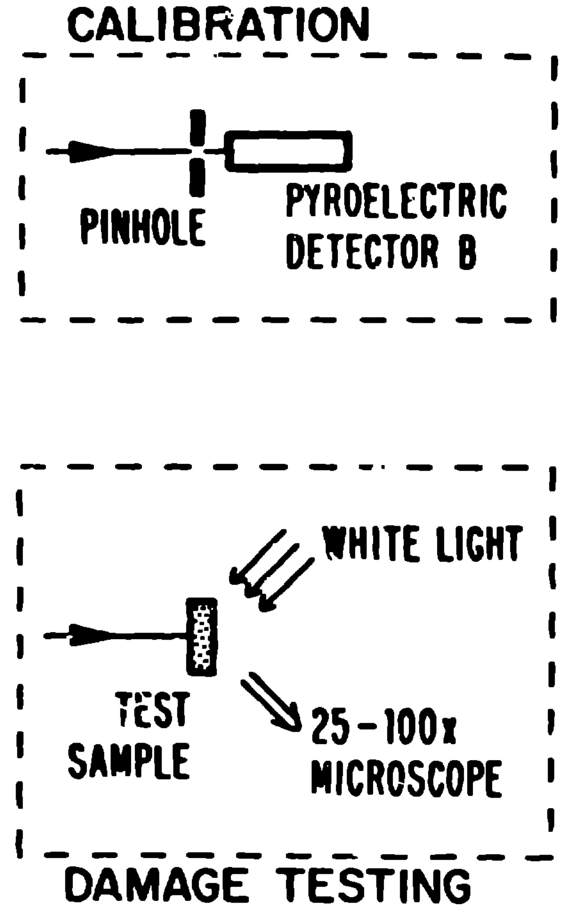
Figure 8. Number of laser shots to obtain damage versus fluence at 308 nm.

Figure 9. Influence of laser preconditioning at 248 nm on the damage threshold behavior.

FIGURE 1.

SCHEMATIC OF UV LASER DAMAGE EXPERIMENT





CALIBRATION – measure ratio: $\frac{\text{fluence B}}{\text{fluence A}}$

TESTING – use fluence A, calibration ratio, and filter factor to continuously monitor fluence B (test fluence)

AP-1-VG-6253

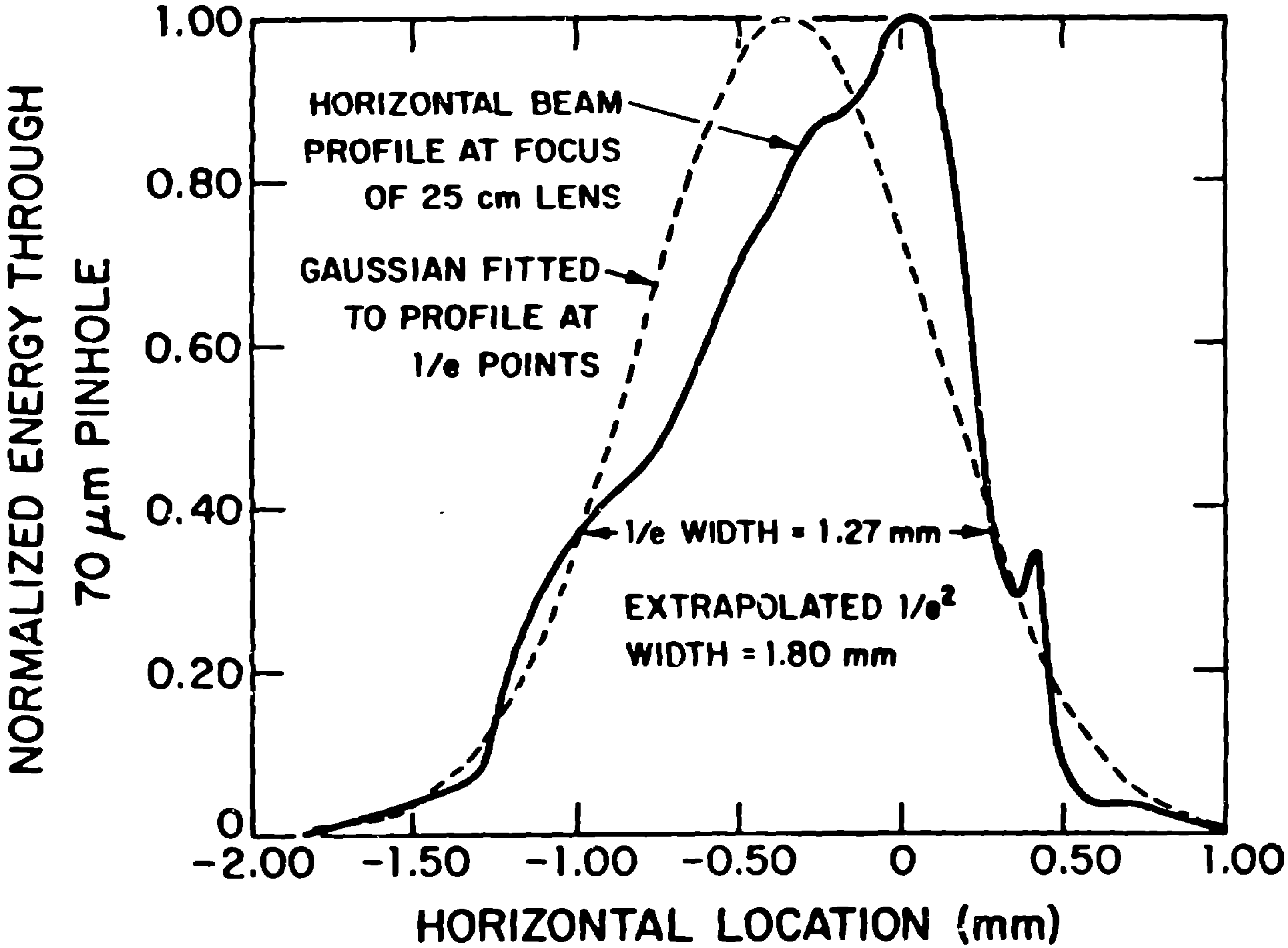


FIGURE 2. SPATIAL PROFILE OF FOCUSSED 248-nm LASER BEAM

AP-1-VG-6251

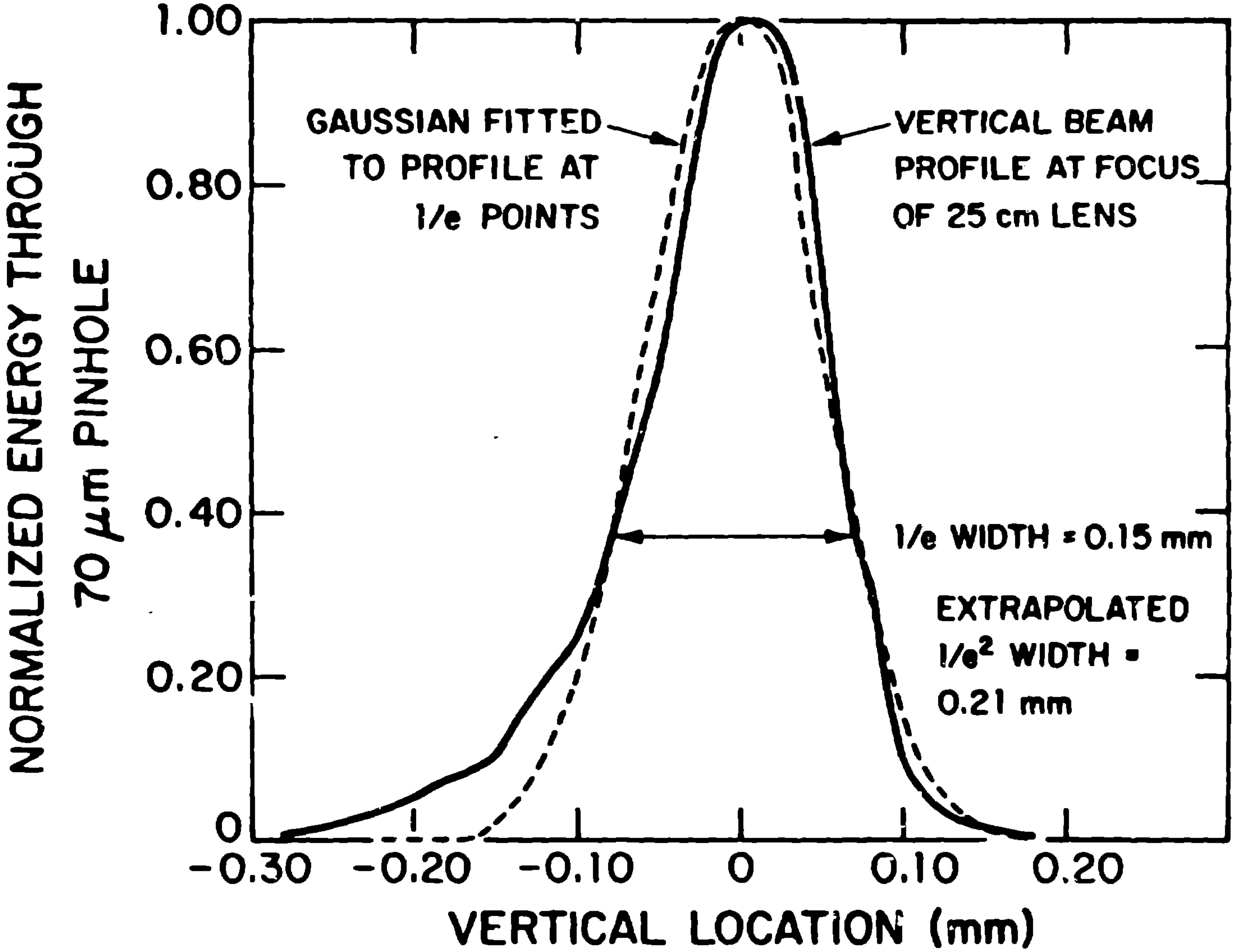


FIGURE 3. SPATIAL PROFILE OF FOCUSSED 248-nm LASER BEAM

AP-1-VG-6252

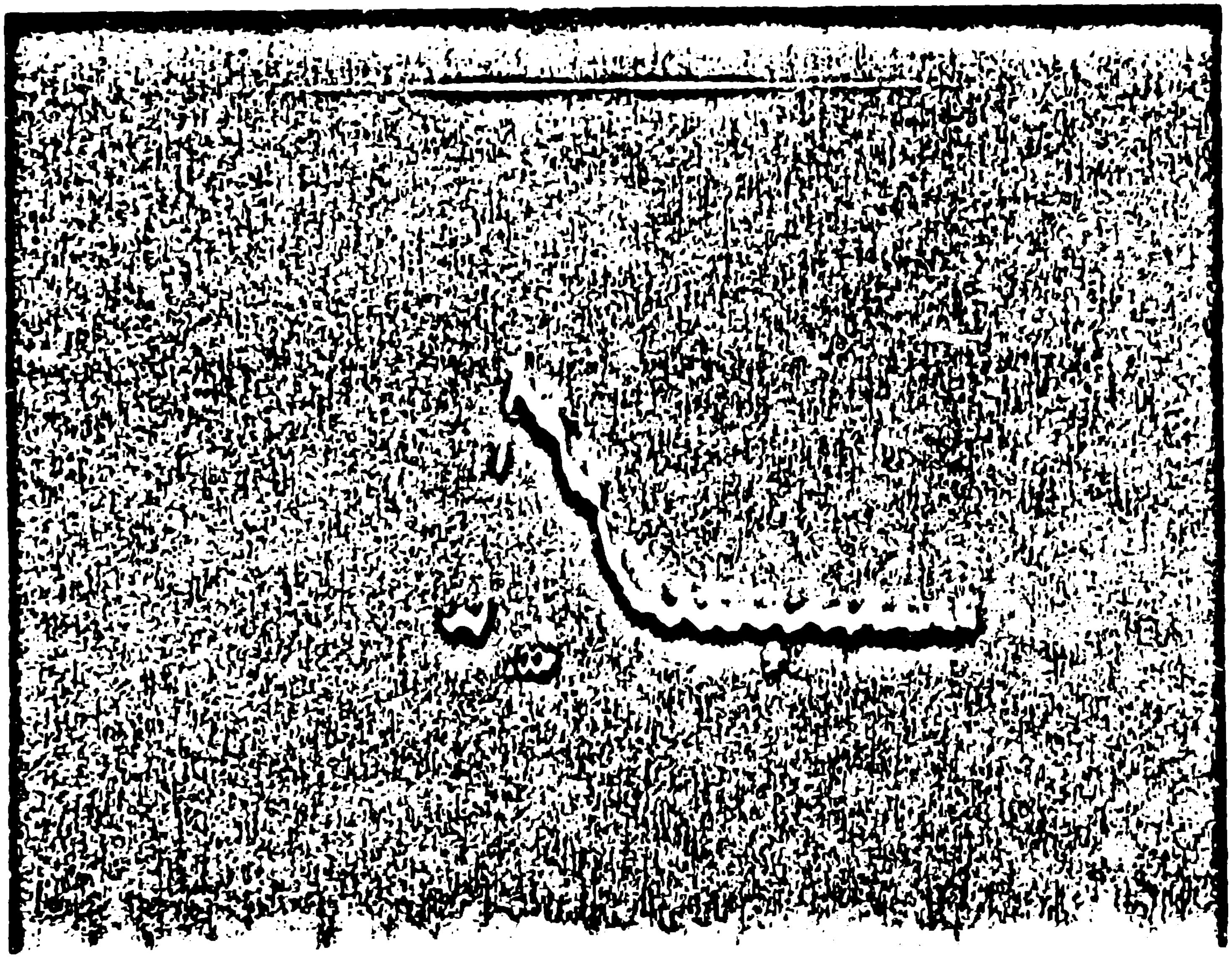


FIGURE 4. OSCILLOGRAM OF THE TEMPORAL DEPENDENCE OF THE LASER PULSE AT 248 nm

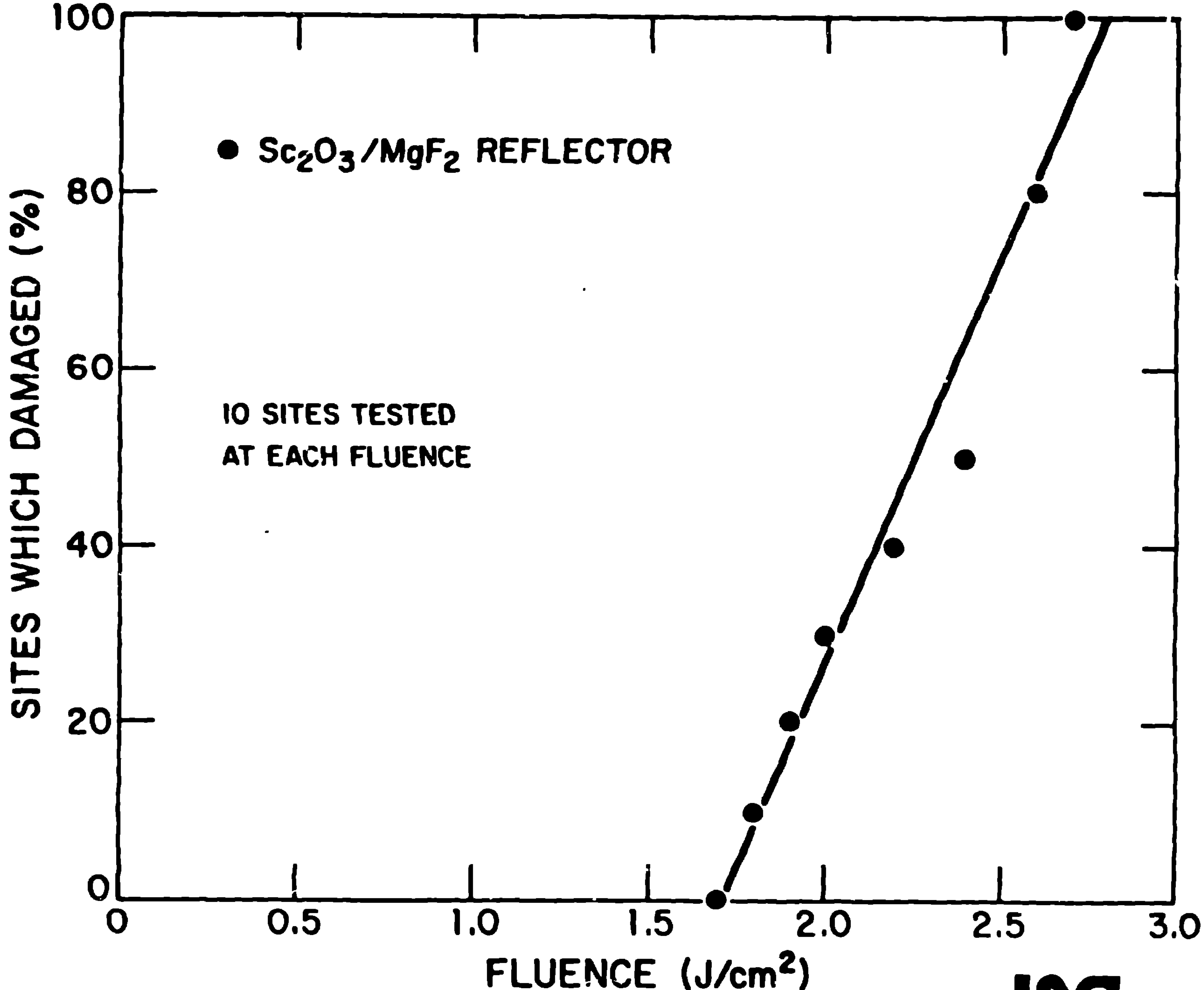


FIGURE 5. DAMAGE VERSUS LASER FLUENCE AT 248 nm

AP-1-VG-6269

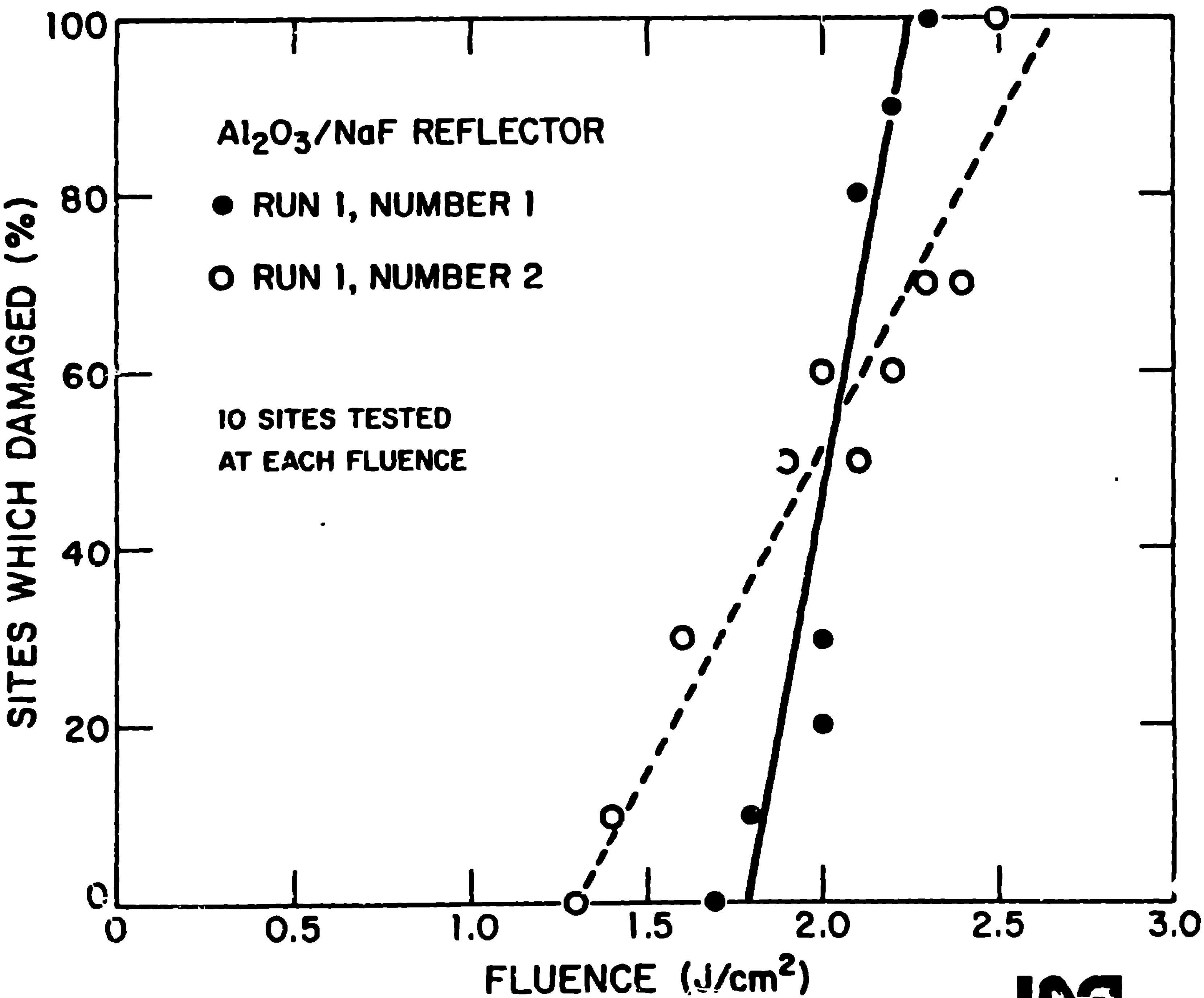
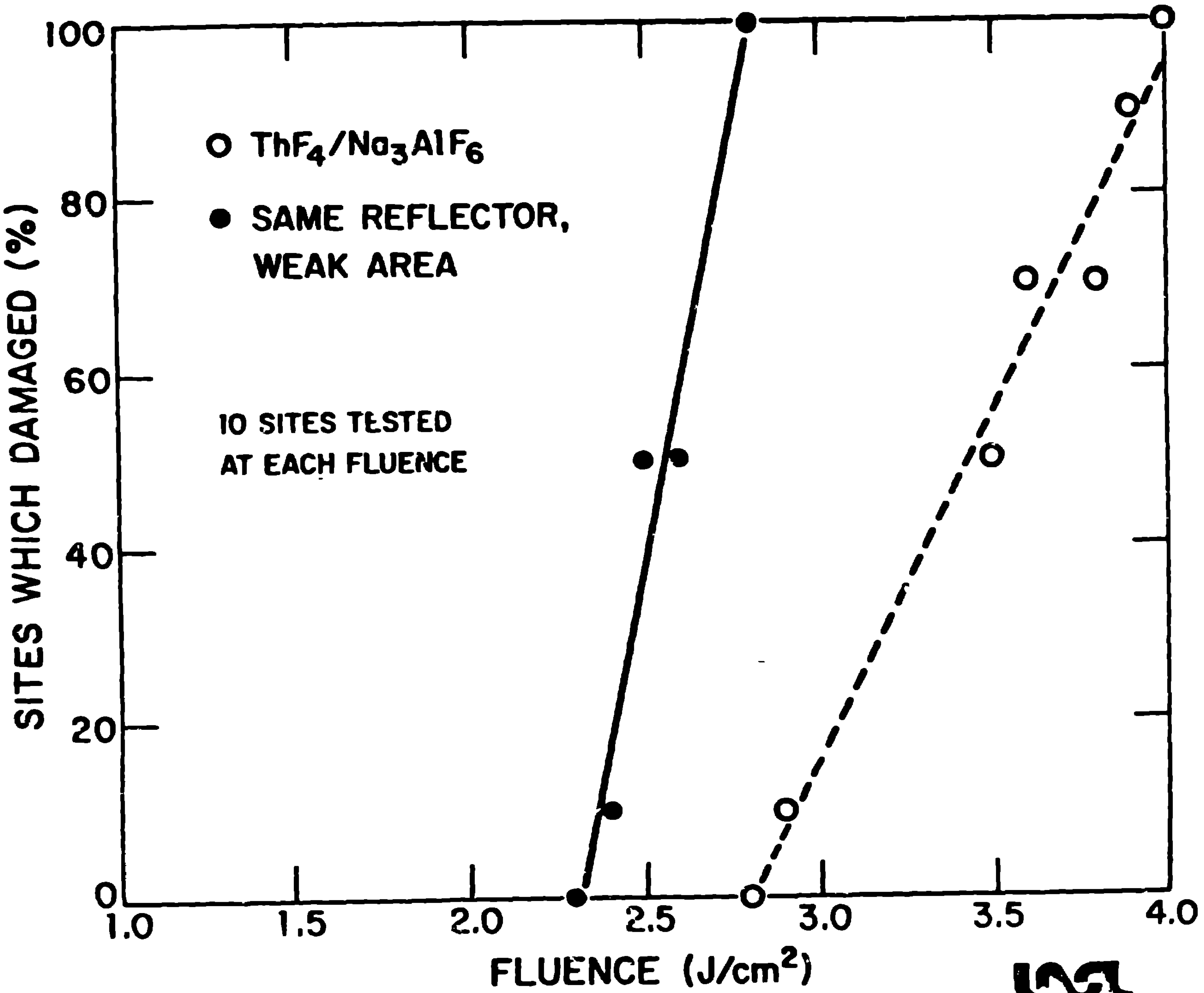


FIGURE 6. DAMAGE VERSUS LASER FLUENCE AT 248 nm

AP-1-VG-6268



AP-1-VG-6266

FIGURE 7. DAMAGE VERSUS LASER FLUENCE AT 248 nm

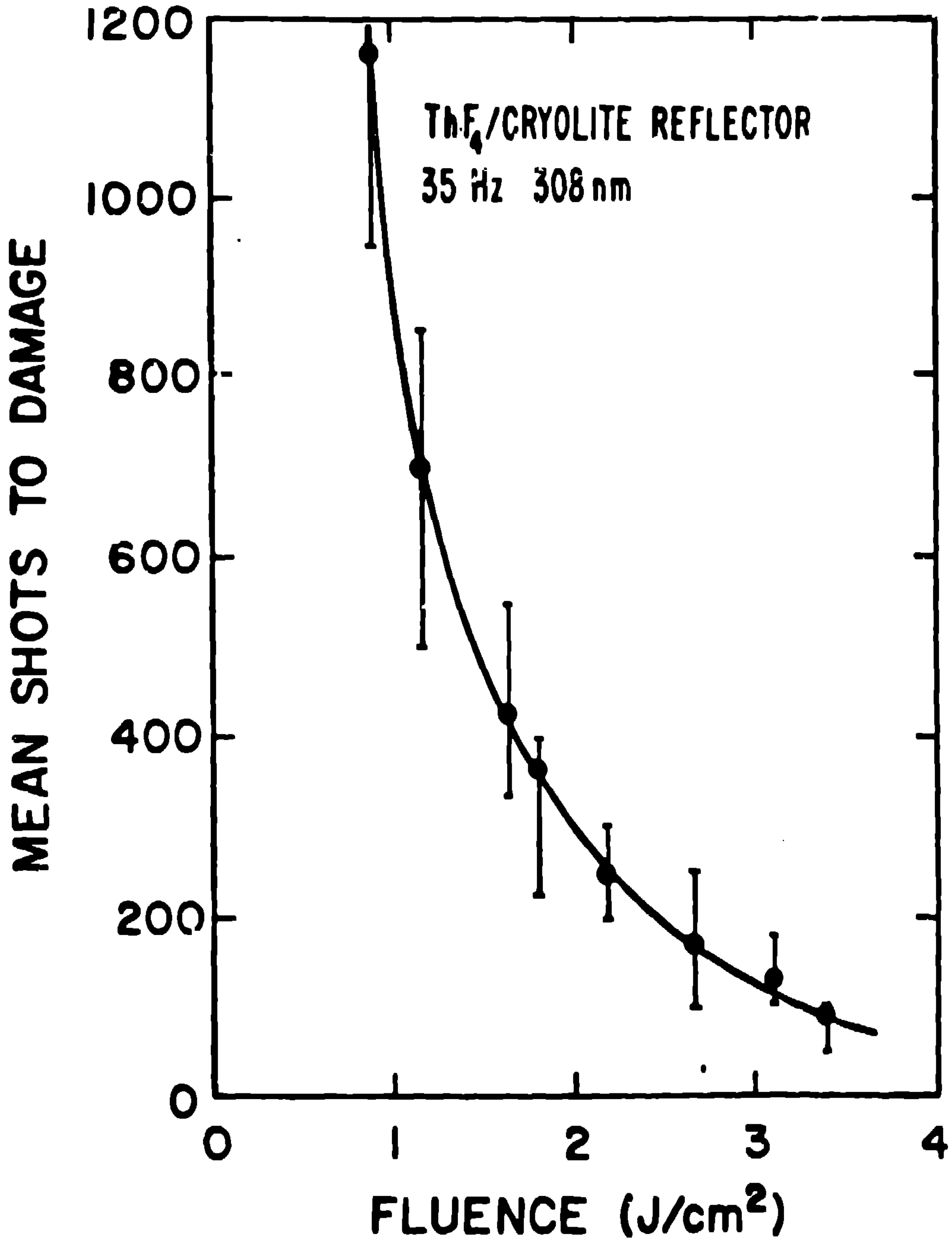


FIGURE 8. NUMBER OF LASER SHOTS AT 308 nm TO OBTAIN DAMAGE VERSUS FLUENCE

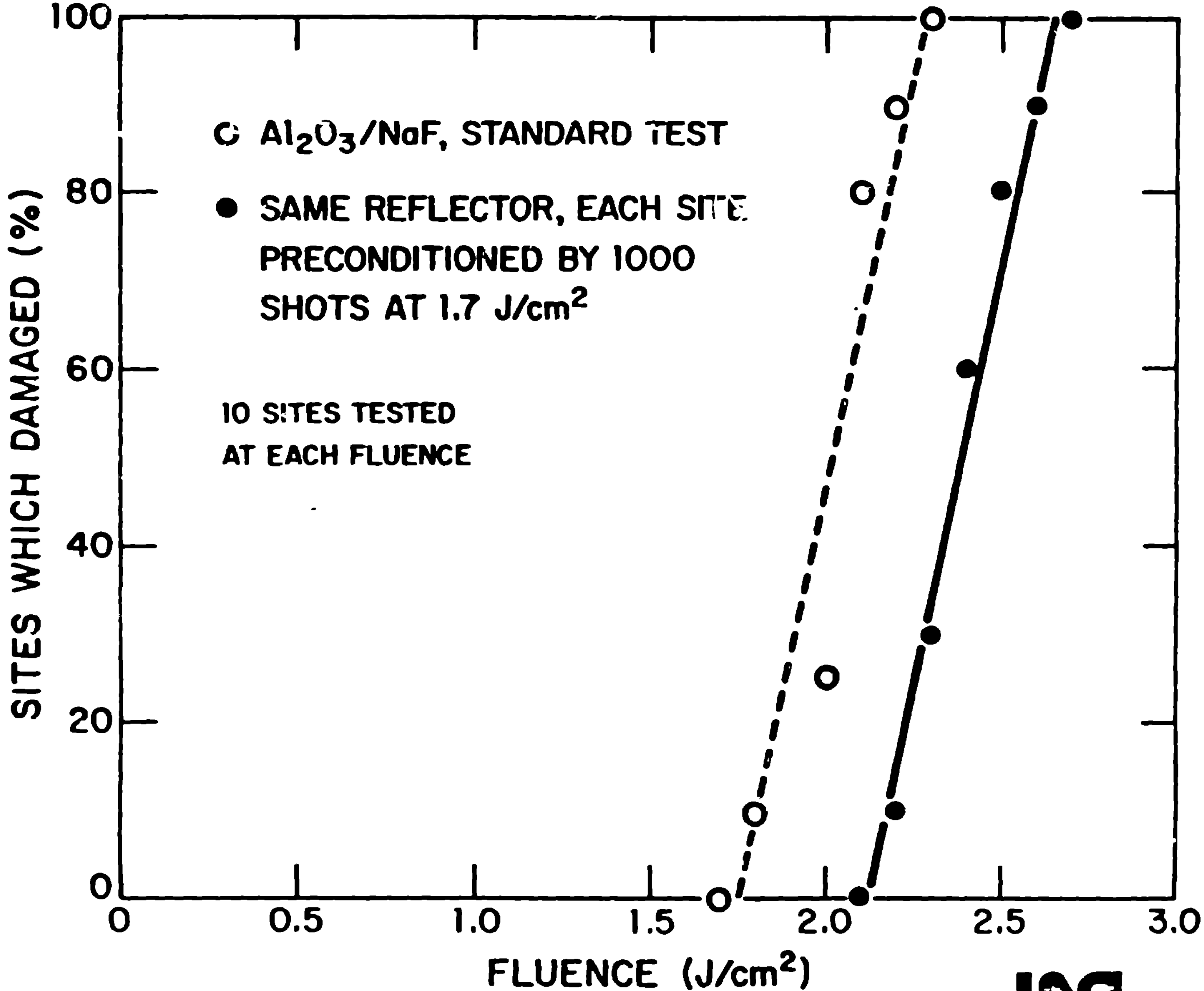


FIGURE 9. INFLUENCE OF LASER PRECONDITIONING AT 248 nm ON THE DAMAGE THRESHOLD BEHAVIOR

AP-1-VG-6267