PULSED HF/DF LASER DAMAGE IN WINDOW MATERIALS*

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Laser damage thresholds are reported for several alkali-halides, alkaline earth fluorides, ZnSe, As_2S_3, sapphire, spinel and quartz at HF (2.7 μm) and DF (3.8 μm) wavelengths. A low pressure, transversly excited, double discharge laser and two different focal length lenses were used. A Gaussian spatial beam distribution was obtained by spatially filtering out high order modes in the far field of an unstable resonator cavity. Sapphire was found to have the highest bulk damage threshold of the materials tested, 100 GW/cm^2 at 2.7 μm (peak intensity on axis). The damage threshold was found to vary as the inverse of the spot diameter which supports the model of Bettis, et. al. (NBS Spec. Publ. 462) The measurements also show that all of the materials evaluated in this effort are surface damage limited when exposed to pulsed HF or DF laser radiation. Evidence concerning the roles of material manufacture, surface finishing and laser irradiation conditioning in the damage process are presented.

Key words: Alkali-halides, alkaline earth fluorides, As_2S_3, DF, electric breakdown, HF, laser damage, quartz, sapphire, spinel, ZnSe.

1. Introduction

Measurements were made of the pulsed laser damage resistance of several candidate window materials at HF and DF laser wavelengths. Of the materials tested sapphire and the alkaline earth fluorides have the highest resistance to laser damage. A major part of the experimentation was devoted to modification and characterization of the HF/DF chemical laser. The damage irradiations were performed with total powers less than 1/10 of the critical powers for self-focusing by tightly focusing the Gaussian laser beam. At 2.7 μm, the HF laser wavelength, a single triangular pulse waveform was used, however, at 3.8 μm, the DF wavelength, a double pulse was obtained because the laser oscillated on several lines simultaneously. The occurance of surface damage set the practical use limit for the materials that were studied and in general there was a considerable effect of laser preconditioning (N on 1 effect) for the surface damage threshold. (1)[1]

2. Experiment

In the damage threshold measurements a transversly excited HF (or DF) pulsed chemical laser was used as the irradiation source. The output wavelength of the HF laser was multiline and centered about 2.7 μm; the output is considerably more dispersed for DF and is centered about 3.8 μm. The only change made in the system to go from HF to DF lasing was to switch from H_2 to D_2 gas. The power supply, laser cavity and all other optics remained the same. The relevant laser parameters and cavity design employed are shown in figure 1.

The laser was manufactured by Lumonics (2) and has been described previously, however, the cavity has been modified as shown in figure 1. The unstable resonator employed a 100% reflector with focal radius of 14 m and an NaCl output coupler of focal radius .8 m. The NaCl lens was uncoated and the 4% reflection from the surface was the only feedback into the cavity. A series of Fresnel rings were observed in the near field of the laser output. These were sensitive to adjustment of either reflector as well as to the size and position of the intracavity aperture. The output traversed the path shown in figure 2. It propagated to a focus 5.76 m "downstream" where a 1.7 mm diameter spatial filter was positioned to block any off axis output modes. This spatially filtered output was then attenuated by two pair of Brewster angle ZnSe slabs; the first pair of which was rotatable to vary the attenuation, and the second was used to assure that the polarization in the beam reaching the target remained constant. After these, bulk attenuators such as quartz, Ge or As_2Se_3 were inserted as necessary. The energy in each pulse was monitored after the attenuators by a calibrated pyroelectric energy meter. At a total distance of 9.0 m from the laser output coupler lens the far field spatial distribution was the Gaussian desired for meaningful damage experiments. At this point the output was focused by a ZnSe meniscus lens (2 different focal length lenses were used) on or inside the samples.

A beam scan of the output of the HF laser at the position of the damage lens with a 0.4 mm aperture gave the data points in figure 3. Each point represents the average of 10 laser shots. The drawn in curve shows a fit of the data to a Gaussian. The $1/e^2$ full width of the Gaussian fit curve is 22.0 mm.

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[1]Figures in brackets indicate literature references at the end of this paper.

A pinhole scan of the laser output was also performed in the focal planes of ZnSe lenses of focal lengths 38.0 and 127 mm. A scan of the focal plane of the 38.0 mm focal length ZnSe lens is shown in figure 4 using a 9.1 µm diameter pinhole. This pinhole was made in thin Al foil by the focused laser output, and its diameter measured in an optical microscope. Each data point represents a single laser shot measured with the pyroelectric energy monitor. The pinhole was moved by a differential micrometer in 12.7 µm intervals. The data has been folded with respect to the maximum. Since the pinhole size was not small compared to the beam size, corrections were made to account for the finite size aperture. (3) It was found by numerical integration (of a circular aperture with a Gaussian weighting function) that the beam width appeared 5% larger using a 9.1 µm diameter pinhole than if the scan had been performed with a much smaller aperture (e.g.: 1 µm). The Gaussian, uncorrected for a finite size aperture is shown as points in figure 4. Even using an aperture as large as one third the $1/e^2$ width causes very little distortion of the observed beam profile.

The temporal pulse waveform was monitored at two points, one before the spatial filter and one after transmission through the sample as shown in figure 2. The HF laser produced a triangular pulse with some irregular spiking which has a full width at half maximum (FWHM) of 175 nsec as shown in figure 5. The HF waveforms were monitored using fast pyroelectric detectors with risetimes of ~1 nsec (Ge photon drag detectors cannot be used at 2.7 µm). The second pulse in the HF waveform is due to ringing in the pyroelectric detector. The DF waveforms were monitored using Ge photon drag detectors and consisted of the double pulse also shown in figure 5. Damage occured on the second part of the pulse as observed by monitoring the waveform distortion upon transmission through the sample. An equivalent pulse width for the DF pulses of 176 nsec was obtained by finding the total normalized pulse area(nsec) and multiplying by the percentage of the area under the second part of the pulse. This is mathematically equivalent to the FWHM obtained for a triangular pulse of the same area. An example of the transmitted pulse waveforms when damage occurred is shown in figure 5 for both HF and DF pulses.

The pulse waveforms for both HF and DF operation were critically dependent on gas mix, pressure, discharge voltage and laser repetition rate. The operating parameters listed in figure 1 were chosen because they resulted in the best HF waveform and this could be reproducibly obtained from day to day. A single DF pulse could not be obtained by simply varying these parameters and, as demonstrated previously, the double pulse DF output is attributed to the multiline nature of the laser oscillation.

3. Calculation

The definition of a damage threshold level of irradiation used in this paper is that flux which produces damage at 50% of the irradiated sites. An example of data is shown in figure 6. The plus or minus values are a measure of the overlap that occurred in all of the samples tested. The 1 on 1 threshold corresponds to one irradiation per site. The n on 1 threshold is where a given site is irradiated by a pulse or pulses with insufficient energy (or intensity) to damage prior to the pulse that produces damage. From the measured energy which caused damage the intensity thresholds were determined in the following way. The energy E in terms of the energy density $\varepsilon(r)$ at the focal plane is given by

$$E = \int_0^\infty \varepsilon(r)\, 2\pi r dr. \tag{1}$$

For the Gaussian beam used in these experiments

$$\varepsilon(r) = \varepsilon_0 e^{-2(r/\omega_0)^2} \tag{2}$$

where ε_0 is the peak on axis energy density and ω_0 is the $1/e^2$ half width point in intensity. Thus,

$$E = \omega_0^2 \varepsilon_0 \pi/2 \tag{3}$$

giving the peak on axis energy density ε_0 as

$$\varepsilon_0 = 2E/\pi\omega_0^2 \tag{4}$$

This equation gives the damage threshold in J/cm^2. To obtain the intensity damage threshold I in watts/cm^2, ε_0 is simply divided by the normalized area, T, of the temporal waveforms; 175 nsec for the HF pulse, and 176 nsec for the DF as discussed previously. Thus

$$I = 2E/(\pi\omega_0^2 T) \tag{5}$$

(Again, this is equivalent to performing the temporal integral for a triangular pulse waveform). The transmission of the ZnSe lenses at both 2.7 and 3.8 µm was measured to be 0.80. The energy E in the previous equation was corrected for this lens transmission in the calculation. In addition for bulk or exit surface damage thresholds the energy was corrected for the front surface reflection. (i.e.: $4n/(1 + n)^2$ of the incident energy is transmitted through an interface where n is the index of refraction of the sample at the appropriate wavelength). In all the samples tested, with the possible exception of quartz at 2.7 µm, absorption was entirely negligible.

4. Data

Table 1 gives the damage thresholds at 2.7 µm (HF) as measured with a 38 mm focal length ZnSe lens (measured $1/e^2$ full width spot size of 27 µm). Also presented is the increase in threshold when a single site was preconditioned by pulses of insufficient energy (energy or intensity) to produce damage (labeled N on 1 increase). The number quoted multiplied by the 1 on 1 damage threshold in J/cm^2 or

GW/cm^2 gives the n on 1 damage threshold. The final two columns give our assessment of whether the bulk damage was due to macroscopic inclusions or was intrinsic. This information was deduced from microscope examination of the damage morphology. For example; if damage occurred at different positions along the focal beam path the damage was deemed inclusion induced. If on the other hand as the sample was traversed perpendicular to the beam all the damage sites appeared similar and lay in a single plane, we deemed the damage to be intrinsic.

Table 1. Bulk thresholds with 27 μm spot size (λ = 2.7 μm)*

Specimen	J/cm^2	GW/cm^2	N on 1 Increase	Inclusions	Intrinsic
NaCl (Harshaw)	3.6×10^3	21	no		X
KCl (Harshaw)	2.9×10^3	16	no		X
KBr (Naval Research Lab)	1.2×10^3	6.6	x 1.12	X	
MgF_2 (Optovac)	13×10^3	75	no		X
BaF_2 (Optovac)	13×10^3	78	no		X
SrF_2 (Optovac)	13×10^3	76	no		X
CaF_2 (1 Raytheon)	15×10^3	88	no		X
CaF_2 (2 Harshaw)	14×10^3	85	no		X
CaF_2 (3 Harshaw)	0.47×10^3	2.8	x 1.7	X	
Sapphire (Optovac)	17×10^3	103	no		X
Spinel (Union Carbide)	12×10^3	68	no		X
Quartz (1 NWC)	8.7×10^3	52	no		X
Quartz (2 General Electric)	1.4×10^3	8.5	x 3.0	X	
ZnSe (Raytheon)	0.46×10^3	2.7	x 1.3	X	
As_2S_3 (Servo)	0.29×10^3	1.7	no	X	
MgF_2 (pressed, NWC)	0.05×10^3	0.29	x 1.7	X	

*$1/e^2$ full width intensity

Table 2 gives bulk damage thresholds in the same format at 2.7 μm for a $1/e^2$ full width spot size of 59 μm (the focal spot size of a 127 mm lens). The greater than signs (>) in front of some of the damage thresholds indicates that there was insufficient laser energy (or intensity) to damage the sample and thus the numbers represent lower limits. No N on 1 data could be obtained for these samples.

Table 2. Bulk thresholds with 59 μm spot size* (λ = 2.7 μm)

Specimen	J/cm^2	GW/cm^2	N on 1 Increase	Inclusions	Intrinsic
NaCl	1.7×10^3	10	no		X
KCl	0.84×10^3	5.0	x 1.1		X
KBr	0.21×10^3	1.2	x 2.9	X	
MgF_2	$>5.8 \times 10^3$	>34	-----		X
BaF_2	6.1×10^3	36	no		X
SrF_2	6.4×10^3	38	no		X
CaF_2 (1)	$>5.8 \times 10^3$	>34	-----		X
CaF_2 (2)	2.9×10^3	17	no		X
Sapphire	$>6.0 \times 10^3$	>35	-----		X
Quartz (1)	3.3×10^3	19	x 1.1		X

*$1/e^2$ full width intensity

Table 3 shows surface damage thresholds again using a 59 μm $1/e^2$ full width focal spot size at the 2.7 μm wavelength. Surface damage was labeled inclusion induced since near threshold more than one damage site was observed within the focal area. The anomalously high threshold quoted for KBr (as compared to the bulk) may have been due to improper focusing.

Table 3. Surface threshold 59 μm spot size* (λ = 2.7 μm)

Specimen	J/cm^2	GW/cm^2	N on 1 Increase	Inclusions	Intrinsic
NaCl	1.3×10^3	7.4	x 1.6	X	
KCl	0.27×10^3	1.6	x 2.7	X	
KBr	0.44×10^3	2.6	x 1.5	X	
MgF_2	1.3×10^3	7.4	x 1.6	X	
BaF_2	0.36×10^3	2.1	x 1.0	X	
SrF_2	0.43×10^3	2.5	x 1.2	X	
CaF_2 (1)	2.6×10^3	15.2	x 1.2	X	
CaF_2 (2)	1.2×10^3	7.0	x 1.3	X	
Sapphire	0.65×10^3	3.8	x 1.2	X	
Quartz (1)	†1.0×10^3	†6.1		X	

*$1/e^2$ full width
†exit surface data

Table 4 shows damage thresholds obtained at 3.8 μm (the DF laser wavelength) at the focus of the 38 mm focal length ZnSe meniscus lens. The $1/e^2$ full width focal spot size of 38 μm was calculated from the measured spot size at the 2.7 μm wavelength and scaled according to wavelength. At the right of the chart the damage threshold in GW/cm^2 at the HF laser wavelength is compared to the damage threshold at the DF wavelength after multiplying by the ratio of the spot sizes, 38 μm/27 μm. The theory of Bettis, et. al. (4) predicts that the damage thresholds should be inversely proportional to the spot diameter. The HF and DF damage thresholds are nearly the same when scaled according to this rule.

Table 4. Bulk thresholds at 3.8 μm

Specimen	38 μm Spot* J/cm^2	38 μm Spot* GW/cm^2	N on 1 Increase	Intensity (GW/cm^2) Scaled to a 27 μm Spot Size	HF Damage Threshold (GW/cm^2) For a 27 μm Spot Size
MgF_2 (single crystal)	7.4×10^3	43	no	60	75
MgF_2 (pressed)	0.12×10^3	0.7	x 1.7	1.0	0.3
Sapphire	13×10^3	72	no	102	103
Spinel	6.9×10^3	39	no	55	68

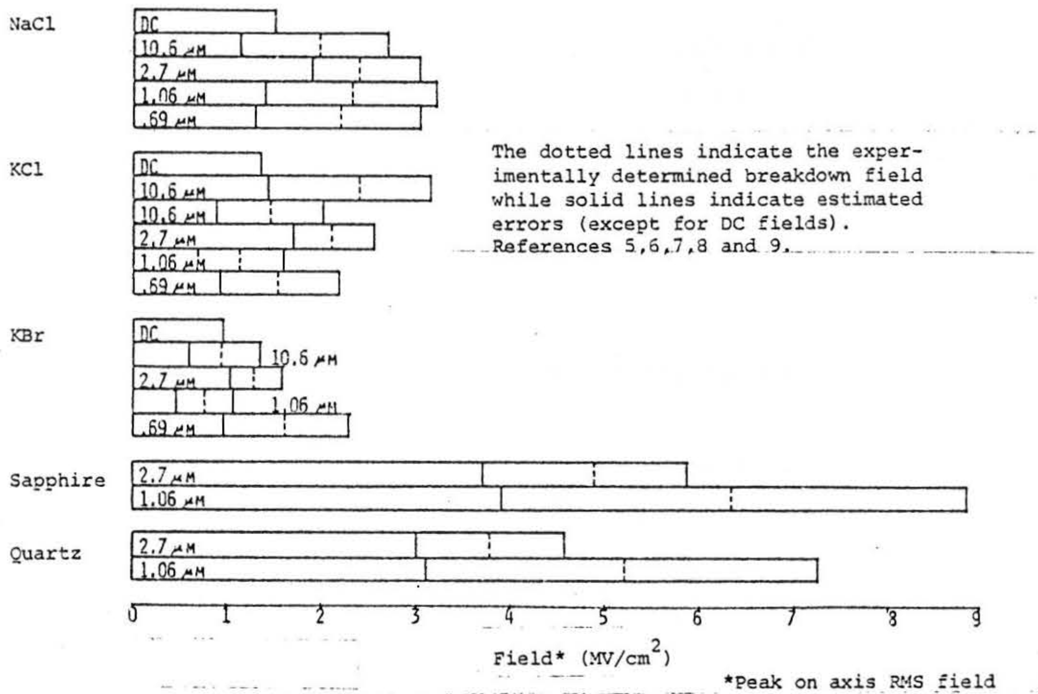
*$1/e^2$ full width intensity

In table 5 we present a bar diagram showing the electric field at which breakdown occurred in the bulk material at various wavelengths including DC. This RMS field was calculated from the intensity damage thresholds, I, and the indices of refraction n, substituted in the equation

$$\text{Field}_{(\text{breakdown})}(\text{Volt/cm}) = 19.41 \sqrt{I(\text{Watts/cm}^2)/n} \tag{6}$$

The 2.7 μm data is from this work. Data at other wavelengths is from the referenced literature. No attempt has been made to reduce the data to account for spot size or pulse width.

Table 5. Breakdown fileds vs wavelengths



The .69 μm Ruby laser data (7,9) used a focal spot diameter (1/e^2 full width in intensity) of approximately 12 μm. The pulse durations were 14 nsec FWHM. At 1.06 μm, the damage threshold appeared spot diameter independent using a spot diameter of from 16 μm to approximately 50 μm. The pulses were about 10 nsec in duration (FWHM). The 2.7 μm and 3.8 μm laser parameters are listed in this paper. The 10.6 μm damage data was taken using a CO_2 laser focal spot diameter of ~100 μm and a pulse length of ~100 nsec. (7,8)

The estimated errors on damage threshold intensities quoted in this paper are ~30%, which implies an uncertainty of ~14% in the fields.

5. Conclusions

The damage data survey presented here shows that sapphire and the alkaline earth fluorides have the highest damage thresholds at both HF and DF laser wavelengths. This is a fortuitous result since both are rugged materials. In particular sapphire is extremely strong and hard.

Bettis, et. al. (4) determined that the damage intensity threshold is proportional to the inverse of the spot diameter. Although their model was developed for surface damage, they attempted to show that their model also applied to bulk damage. From this we expect that the ratio of the damage intensities for the 27 μm and 59 μm spot diameters should be 2.2. The average value of this ratio determined by this experiment is 2.5 which is in good agreement.

Using this spot diameter scaling and extending it to the 3.8 μm wavelength our DF damage thresholds are nearly the same as the HF thresholds (see table 4). Damage thresholds for some of the materials tested were obtained previously (2) at 3.8 μm using a 130 μm spot diameter (1/e^2 full width). Scaling these values with spot diameter according to reference 4 also gives excellent agreement with the 2.8 μm data presented here. Thus it appears that the damage thresholds are insensitive to wavelength between 2.7 μm and 3.8 μm. We should note, however, that due to the time structure of the DF laser pulse (see figure 5) this data is less reliable.

As has been noted at other wavelengths optical materials are limited by surface damage thresholds. Our data of table 3 confirms this at 2.7 μm. The large laser conditioning effect (n on 1), the low thresholds relative to the bulk values, and microscopic examination show that surface damage is defect limited. Here again the fact that sapphire and the alkaline earth fluorides have high thresholds is helpful since we expect that with careful surface preparation the damage thresholds of these hard materials can be substantially increased. (10)

It is interesting to note that to within experimental error the raw data of table 5 shows a damage field independent of wavelength.

6. References

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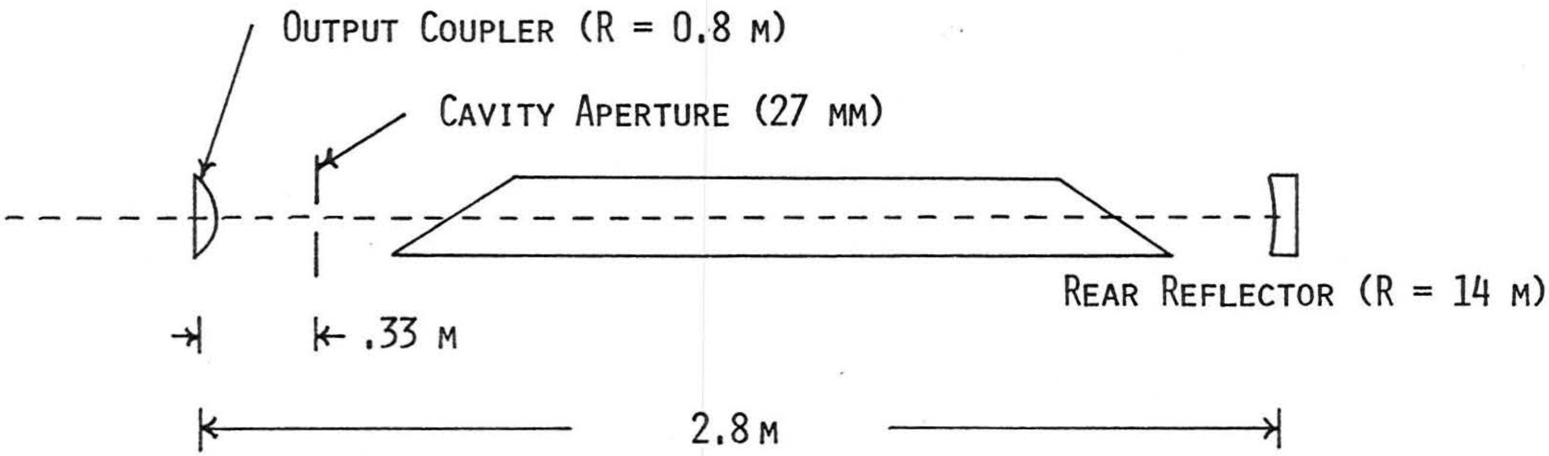
Figure Captions for "Pulsed HF/DF Laser Damage in Window Material

1. HF/DF Laser Cavity
2. Experimental Arrengement
3. HF Laser Spatial Profile (Prior to ZnSe Lens)
4. Pinhole Scan of Focal Plane
5. Pulse Waveforms
6. Threshold Definition

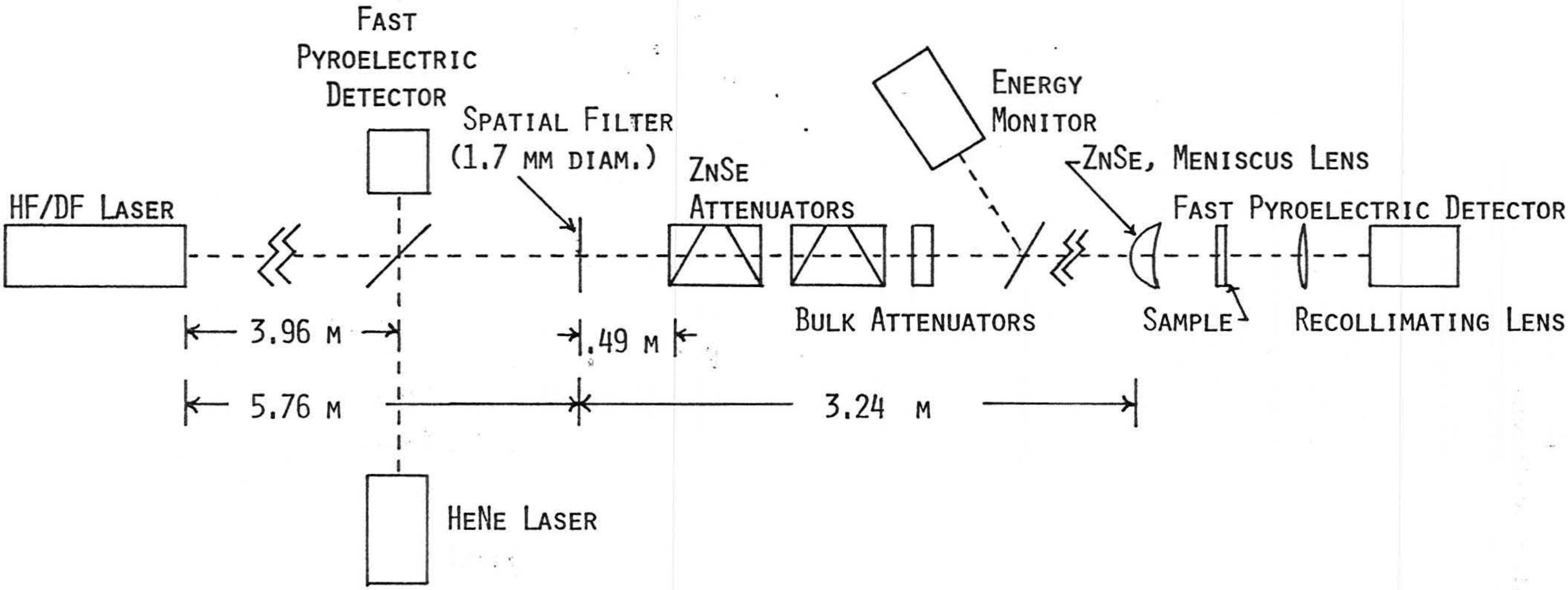
HF/DF LASER CAVITY

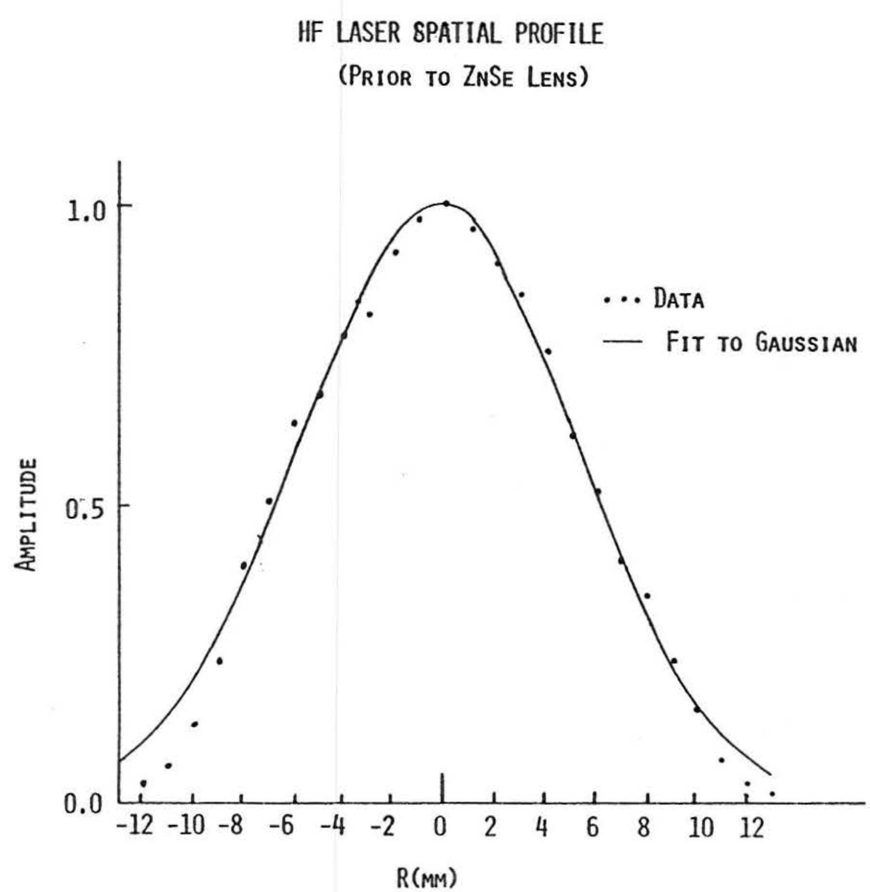


LASER PARAMETERS:	PRESSURE		60 TORR
	DISCHARGE VOLTAGE		72 KV
	GAS MIX:	H_2/D_2	13 LITERS/MIN*
		SF_6	2.2 LITERS/MIN*
		HE	2.6 LITERS/MIN*
	WAVELENGTH:	HF	2.7 μM
		DF	3.8 μM

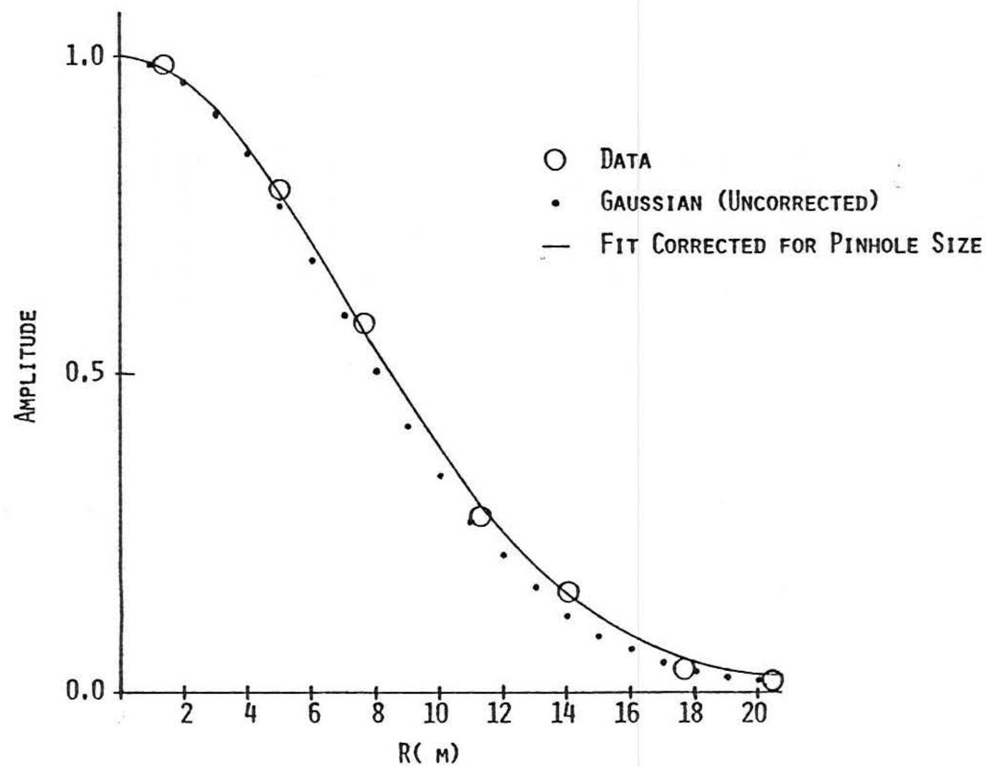
*(STP)

EXPERIMENTAL ARRANGEMENT



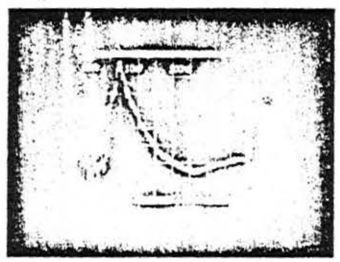

HF LASER SPATIAL PROFILE
(PRIOR TO ZNSE LENS)
• • • DATA
— FIT TO GAUSSIAN
AMPLITUDE
1.0
0.5
0.0
-12 -10 -8 -6 -4 -2 0 2 4 6 8 10 12
R(MM)

PINHOLE SCAN OF FOCAL PLANE

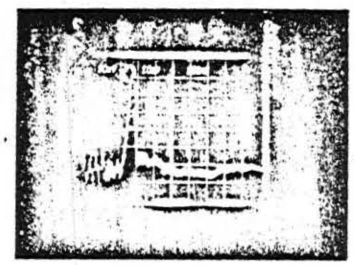


PULSE WAVEFORMS

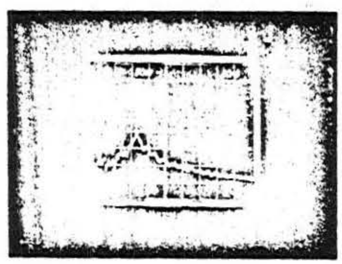
HF INPUT PULSE



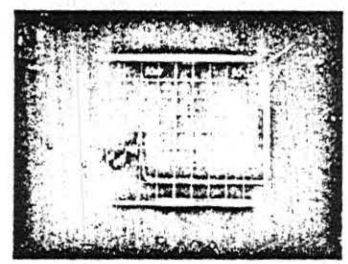
HF DAMAGE



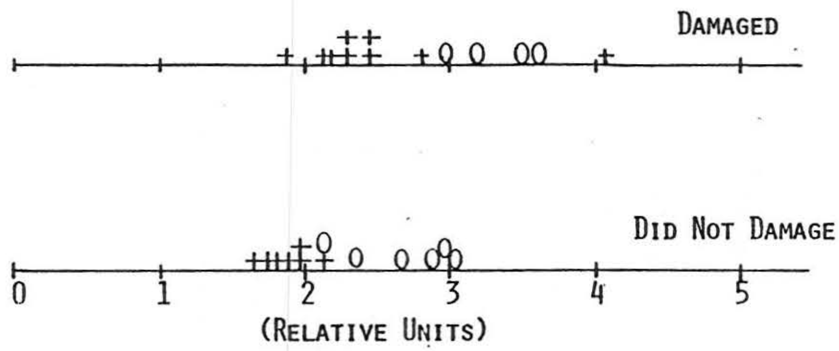
DF INPUT PULSE



DF DAMAGE



THRESHOLD DEFINITION



+ 1-ON-1 THRESHOLD 2.0 ± 0.2

O N-ON-1 THRESHOLD 3.0 ± 0.1