10.6 μm LASER DAMAGE IN COATINGS CONTAINING As_2S_3 AND As_2Se_3 †

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The laser damage properties of 10.6 μm coatings containing As_2S_3 and As_2Se_3 are reported. A TEM_{00} mode CO_2 TEA laser with an intracavity CW CO_2 discharge section was the irradiation source in these experiments. This enabled the study to include a test of the role of mode locked pulses on the damage thresholds of the sample coatings.

Coatings containing As_2S_3 were damaged only after an incandescence or spark was observed during the irradiation. There was no difference in the intensity threshold, ~300 MW/cm^2 (peak intensity on axis) for damage in half wave or full wave thick As_2S_3 coatings, with or without mode locked pulses in the laser waveform. However, evidence for laser irradiation conditioning was found in certain areas of the As_2S_3 coatings.

Two different thresholds were observed for coatings containing As_2Se_3; one where a spark was observed and a large damage site produced and a second, at ~30% lower intensity which produced a very small damage site even though no incandescence was detected. The latter occurred at ~12 MW/cm^2 in both the full and half wave As_2Se_3 coatings. Otherwise, the qualitative behavior of the threshold was similar to that of the As_2S_3 coatings.

The intensity of threshold for damage to As_2S_3/KCl/As_2S_3 anti-reflection coatings on KCl substrates was as high as 430 MW/cm^2 and did not depend on the presence of mode locked pulses. Three layer AR coatings containing As_2Se_3 damage at ~20 MW/cm^2. The relationships between the coating damage thresholds and microstructure, design and measured absorption are discussed.

Key words: As_2S_3; As_2Se_3; coating damage; coating design; defects; laser damage.

1. Introduction

Thin film coatings are often the most easily damaged components of an infrared laser system. The 10.6 μm, pulsed damage thresholds of films containing As_2S_3 and As_2Se_3 are typically ~325 MW/cm^2 and ~15 MW/cm^2 respectively. These thresholds were found not to depend significantly on the coating design, the details of the laser pulse waveform or the sequence of the irradiation employed. Layered coating designs used to reduce coating absorption by redistributing the fields in the layers (1)[1] do not significantly reduce the energy absorbed per unit mass of absorptive material. Thus the heating of the coating on exposure to optical irradiation and any failure properties governed by uniform absorption should be independent of the coating design. However, an examination of the morphology of the damage sites reveals that microscopic defects or inclusions are responsible for the failure of the coatings studied to withstand intense laser irradiation. Initial "survival curve" data is consistent with this model since it rules out uniform linear absorption as the dominant pulse damage interaction.

2. Experimental

The experiment is shown schematically in figure 1 and the properties of the laser system are listed in table I. The low pressure CW discharge in the TEA laser cavity was used to restrict oscillation to a single longitudinal mode (2) and thus to supress mode locked pulses in the output. Figure 2 shows the two waveforms used in these experiments.

Coatings were prepared at the Hughes Research Laboratory, (HRL), in Malibu, California, on single and poly-crystalline KCl substrates. The substrate surfaces were polished and etched. The three layer coatings were designed as anti-reflection coatings in the manner discussed later in this paper. Half

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

and full wave layers were studied to test the role of coating design in determining the damage threshold.

The occurrence of damage was detected by an observer viewing the irradiated site through a long working distance microscope. Both white light and HeNe laser light scattering were used with comparable results. The coatings which contained As_2Se_3 often damaged with no visible sparks or incandescence at fluxes 30% less than required to produce a spark. However, As_2S_3 coatings only damaged after the irradiation produced a spark.

Both 1 on 1 and N on 1 irradiation sequences (3) were used but only the uncoated substrate showed a significant irradiation conditioning effect. The irradiated sites were inspected with visible microscopy to determine the damage site morphology. Normal incidence reflected, bright field transmitted, and phase contrast transmitted illumination were used to maximize the effectiveness of the morphological examinations.

Table 1. CO_2 laser parameters & performance data.

TEA Section:	
Discharge Length (Double Rogowski Electrodes)	41 cm.
Energy Storage Capacitance	0.08 µf
Flow Rate Ratio ($He:N_2:CO_2$)	8.7:2.4:2.6 ℓ/min.
Typical Input Energy into Pulsed Section	21 J
CW Section:	
Discharge Length	104 cm.
Gas Mixture	86%:7%:7%
Gas Pressure Flowing gas, water cooled	3 Torr
Mirrors	100% R - flat/Si 80% R - 10 meter/Ge
Brewster Windows	KCl
Intracavity Aperture Diameter for TEM_{00} Mode	8 mm.
Total Cavity Length	307 cm.
Maximum TEM_{00} Energy Available at the Target	30 mJ
Typical Width of Pulse (FWHM)	180 nsec

3. Measured Damage Thresholds

The damage thresholds obtained in these experiments are listed in table 2. The values for coating absorption were measured by H.R.L. as part of the coating characterization. Clearly coatings containing As_2S_3 can withstand 20 times the flux that will damage one containing As_2Se_3. The data also demonstrate that the damage flux for each type of coating does not depend strongly on the coating design. The last entry for an $As_2S_3/KCl/As_2S_3$ three layer coating represents data for a poor quality coating. No significant laser conditioning (N on 1) effect was observed except for the uncoated KCl substrate surface. In addition, the threshold for all the listed samples did not depend on which pulse waveform was used (fig. 2).

Table 2. Damage thresholds for As_2S_3 and As_2Se_3 containing coatings.

Material	Absorption % / Coating	Energy Density* (J/cm^2)	Power Density* (MW/cm^2)
Surface of KCl single crystal (N on 1)		$\boxed{314}$ 171 157 (329)	871 (1720)
$As_2Se_3 - \lambda/2$ (N on 1)	0.02	2.40 2.29 (2.40)	12.7 (12.7)
$As_2Se_3 - \lambda$ (N on 1)	0.018	2.60 2.43 (3.03)	13.5 (15.8)
$As_2Se_3/KCl/As_2Se_3$ (N on 1)	0.03	3.85 3.62 (3.85)	20.1 (20.1)
$As_2Se_3/KCl/As_2Se_3$ (N on 1)	0.02	3.3 3.10 (3.3)	17.2 (17.2)

196 nsec pulse

Table 2. Damage Thresholds... (continued)

Material	Absorption % / Coating	Energy Density* (J/cm^2)	Power Density* (MW/cm^2)
$As_2S_3 - \lambda/2$ (N on 1)	0.067	60 60 (88)	311 (462)
$As_2S_3 - \lambda$ (N on 1)	0.12	56 53 (56)	293 (293)
†$As_2S_3/KCl/As_2S_3$ (N on 1)	0.029	83 98 (120)	433 (564)
†$As_2S_3/KCl/As_2S_3$ (N on 1)	0.045	8.1 9.6 (8.1)	42 (42)

*The thresholds quoted are the peak on axis values of the indicated quantity, and $2W_o$ = 125 μm is the diameter at the focus measured to $1/e^2$ of the on axis intensity. The pulse duration was 180 nsec.

†The three layer coatings are anti-reflection coatings at 10.6 μm.

4. Coating Design

Figure 3 shows the field distributions in the half wave and the three layer anti-reflection coatings containing As_2S_3 which were studied. If the As_2S_3 layers are responsible for absorption in the coating then the three layer design will have less absorption. This is so because there is less of the absorptive material exposed to the light field in the three layer design. However, in the three layer design less material is responsible for proportionally less absorption. Thus, the energy absorbed per unit mass of coating material is unchanged and each coating will be heated to the same temperature by the same light flux. By this argument it is possible to interpret the measured damage thresholds as being caused by uniform linear absorption. The presence of defects or inclusions in the coatings will also give rise to a damage process which is independent of coating design and as shown in the next sections is in fact the principal failure mechanism.

5. Morphology of Coating Damage and Coating Uniformity

In figures 4 through 9 several representative examples of coating damage morphology are shown. Figures 4 through 9 show that in the irradiated area (~125 μm in dia.) there were one or more randomly located sites which were easily damaged. The crazing in figures 8 and 9 is due to the polycrystalline nature of the substrate on which the coating was placed. Figure 9 shows that when a spark was produced on an As_2Se_3 coatings, a significant portion of the coating was completely removed. In this context it is important to note the value of using more than one type of microscope illumination to examine laser damage.

In figure 5, a case of spark damage to As_2Se_3 coating, there are interference "ripples" with spacing λ/n KCl similar to those described by Temple and Soileau.(4) These are due to light scattered from a defect in interfering with the incoming laser beam. The coating to substrate interface is an "exit" surface in the same sense as in reference 4 and the "ripples" are found, as expected, on the KCl substrate surface.

Figure 10 is a plot of the damage threshold as a function of position on an As_2Se_3 coating. This data demonstrates the point-to-point non-uniformity of the coating and is additional evidence for a damage mechanism dominated by the presence of defects or inclusions.

6. Survival Curve Data

A survival curve is a statistical means to examine the mechanism causing laser damage.(5) A uniform intensity pulse is used to irradiate several sites on or in a sample and the statistics of the period of time each site can survive this irradiation is used to determine the damaging interaction. If the dominant interaction is uniform linear absorption, all sites will be identical and all will survive exactly the same period of time. Figure 11 shows the distortion of a "flat-topped" laser pulse which occurs when three different sites on a ZnSe containing coatings were damaged. Clearly the three sites survive different times and so these data rule out a significant role for uniform linear absorption in the pulsed laser damage process for such a coating.

More extensive survival curve studies for bulk material and coatings are planned. The initial data presented here shows the potential for this technique to sort out damage mechanisms.

7. Summary and Conclusions

The principal results of this work are summarized in table 3. Included is data from 1976 on ZnSe containing coatings (6) which show that the best currently available coatings are those containing As_2S_3 or ZnSe. The damaging interaction in all the coatings tested is determined by the presence of microscopic defects or inclusions

Table 3. Summary of Coating Damage Thresholds

KCl Substrate Surface	1000 MW/cm^2
Coatings on KCl Coatings:	
As_2Se_3	15 MW/cm^2
As_2S_3	325 MW/cm^2
ZnSe	450 MW/cm^2

Cause of Coating Failure:
Mechanical Defects and/or Inclusions

8. Acknowledgments

We wish to acknowledge the assistance of J. Menders, A. Stewart, K. Moravvej and M. Harvey in the damage measurements.

9. References

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(2) A. Gondhalekar, N.R. Heckenberg and E. Holzhauer, J. Quantum Electr. QE-11, 3 (1975).

(3) M. Bass and K.M. Leung, J. Quantum Electr. QE-12, 82 (1976).

(4) P.A. Temple and M.J. Soileau, NBS Special Publication 462, p. 371.

(5) D. Milam, R.A. Bradbury, R.H. Picard and M. Bass, NBS Special Publication 414, p. 169.

(6) D. Zuccaro, M. Braunstein, M. Bass, K. Leung and C. Tang, "Status Report of 3 Layer 10.6 µm A-R Coatings on KCl", Proceedings of Infrared Laser Window Materials Meeting, July 12, 1976, Boulder, Colorado.

Figure Captions for "10.6 μm Laser Damage in Coatings Containing..."

1. Schematic of the Equipment

2. Temporal waveforms used in the damage measurements.

3. Electric field distributions in coatings containing As_2S_3. The electric field squared is normalized to the electric field in the substrate.

4. Damage near threshold to a λ/2 As_2S_3 coating on KCl at 10.6 μm.

5. Damage near threshold to a λ/2 As_2Se_3 coating on KCl to 10.6 μm (Upper photo is for damage with no spark and lower is a case where a spark was observed.

6. Damage near threshold to a λ As_2Se_3 coating on KCl at 10.6 μm.

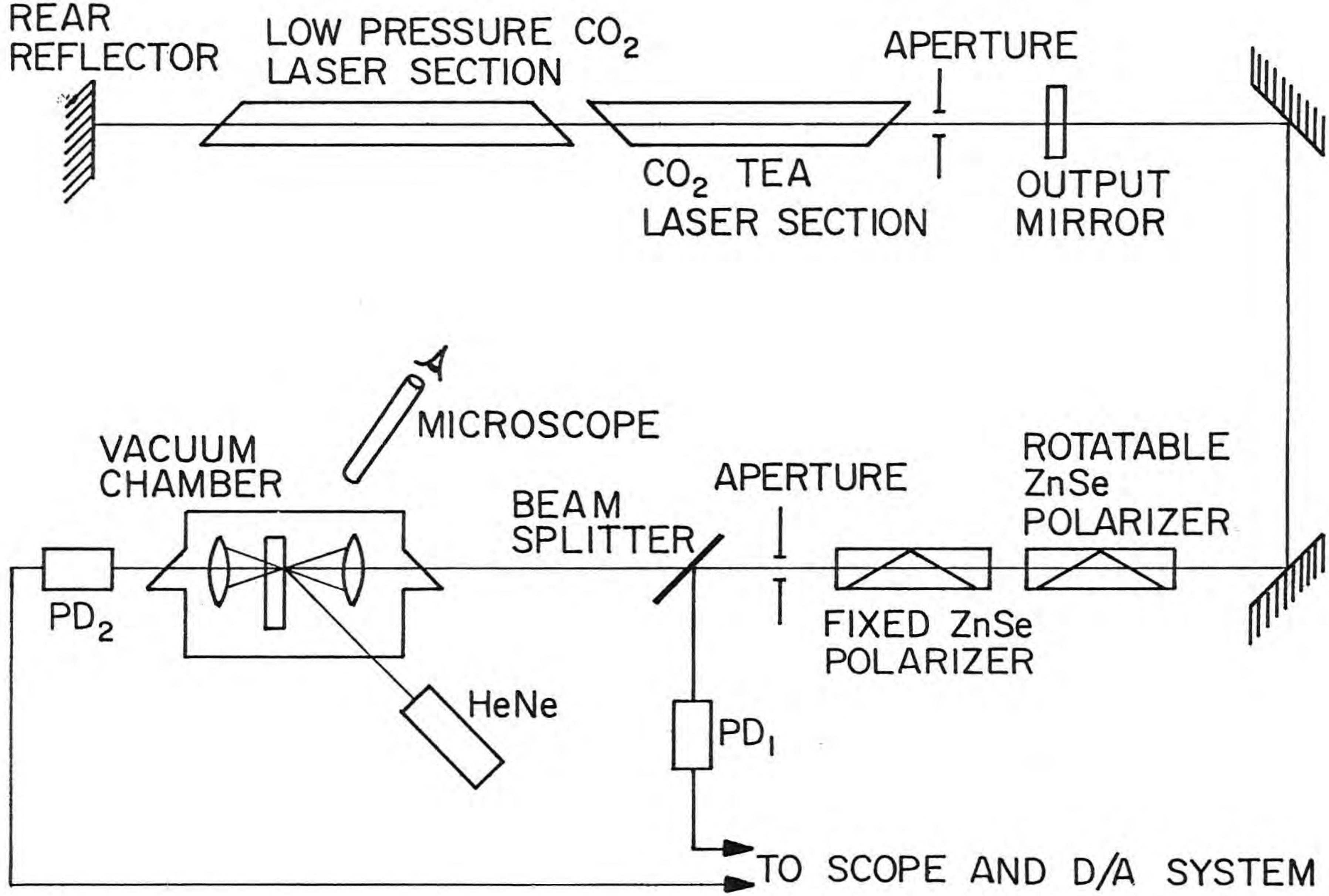
7. Damage near threshold to an As_2Se_3/KCl/As_2Se_3 10.6 μm anti-reflection coating on KCl.

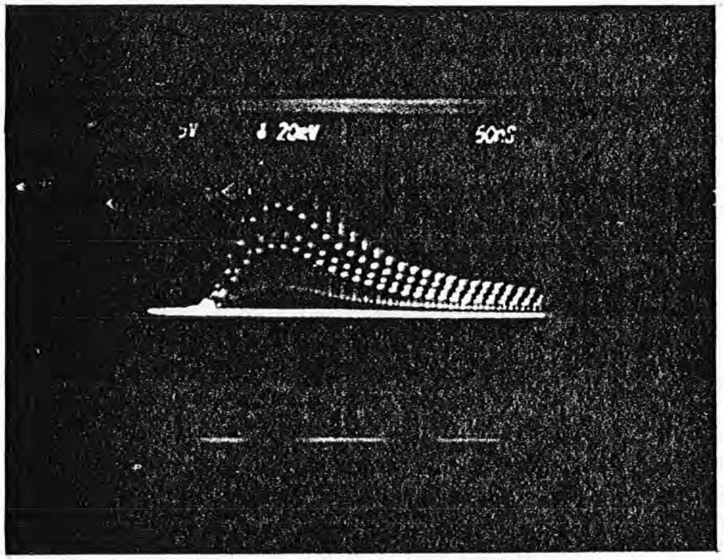
8. Damage near threshold to an As_2Se_3/KCl/As_2Se_3 10.6 μm anti-reflection coating on polycrystalline KCl when no spark was produced during the irradiation.

9. Damage to an As_2Se_3/KCl/As_2Se_3 10.6 μm anti-reflection coating on polycrystalline KCl when a spark was produced during the irradiation.

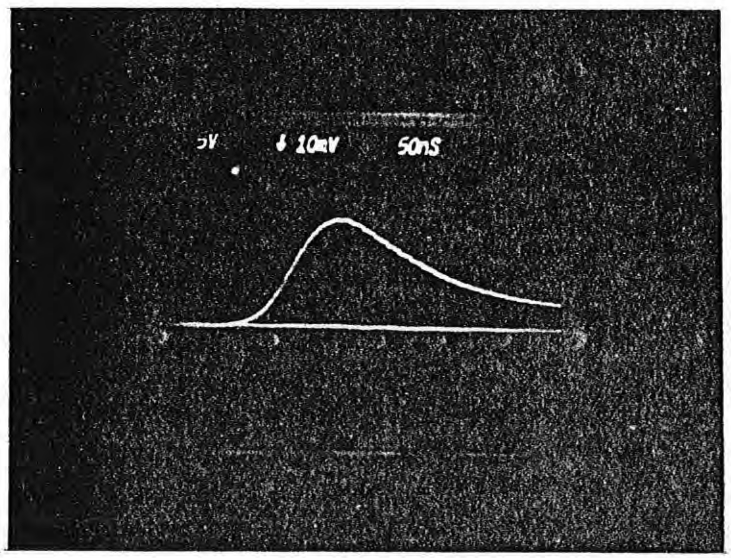
10. Laser damage threshold versus position on an As_2Se_3 coating. This demonstrated the site-to-site non-uniformity of the coating.

11. Transmitted waveform distortion due to 10.6 μm laser induced damage in a AnSe/KCl/AnSe anti-reflection coating on KCl. The incident waveform was a flat topped pulse ~30 μsec long. This demonstrates the site-to-site non-uniformity of the coating.


REAR
REFLECTOR
LOW PRESSURE CO_2
LASER SECTION
APERTURE
CO_2 TEA
LASER SECTION
OUTPUT
MIRROR
MICROSCOPE
VACUUM
CHAMBER
BEAM
SPLITTER
APERTURE
ROTATABLE
ZnSe
POLARIZER
PD_2
FIXED ZnSe
POLARIZER
HeNe
PD_1
TO SCOPE AND D/A SYSTEM

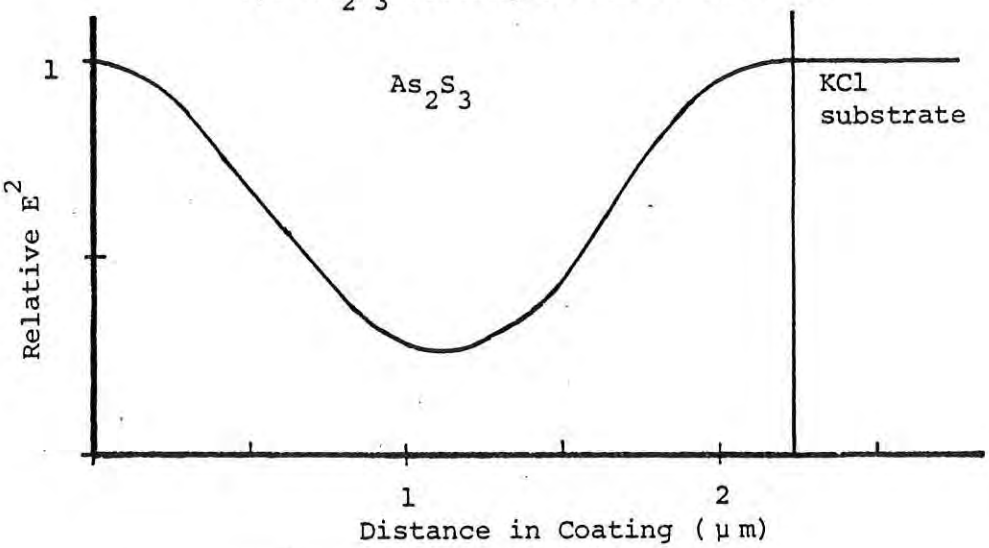


MODE-LOCKED PULSED LASER OUTPUT

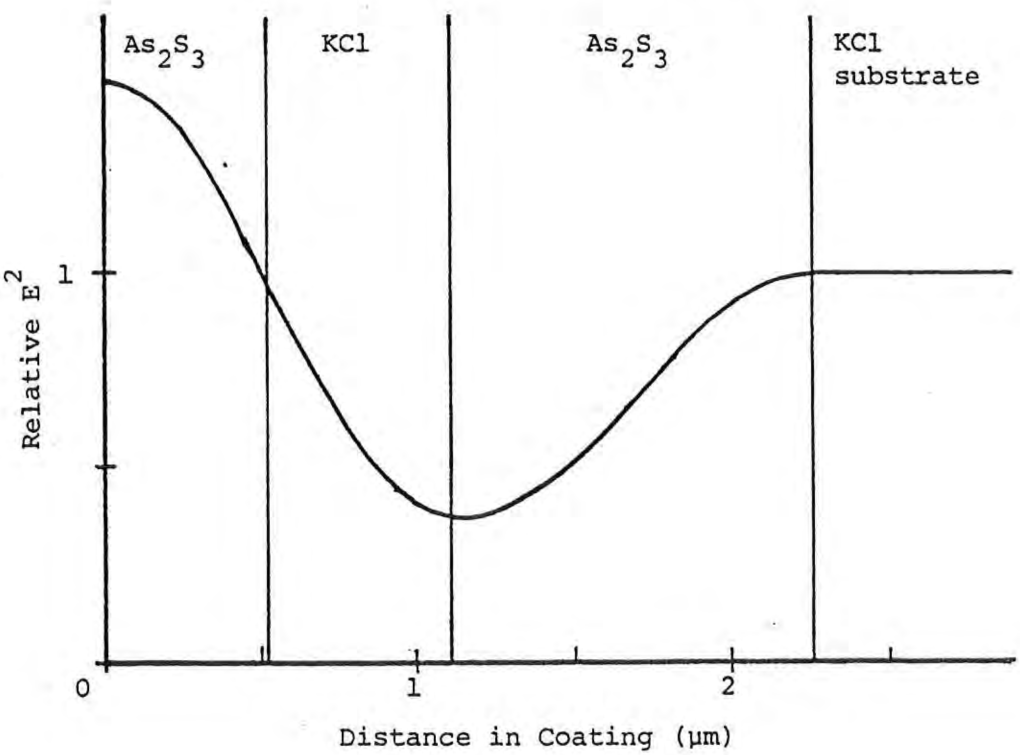


TEMPORAL PROFILE OF PULSED LASER OUTPUT
USING CW SECTION

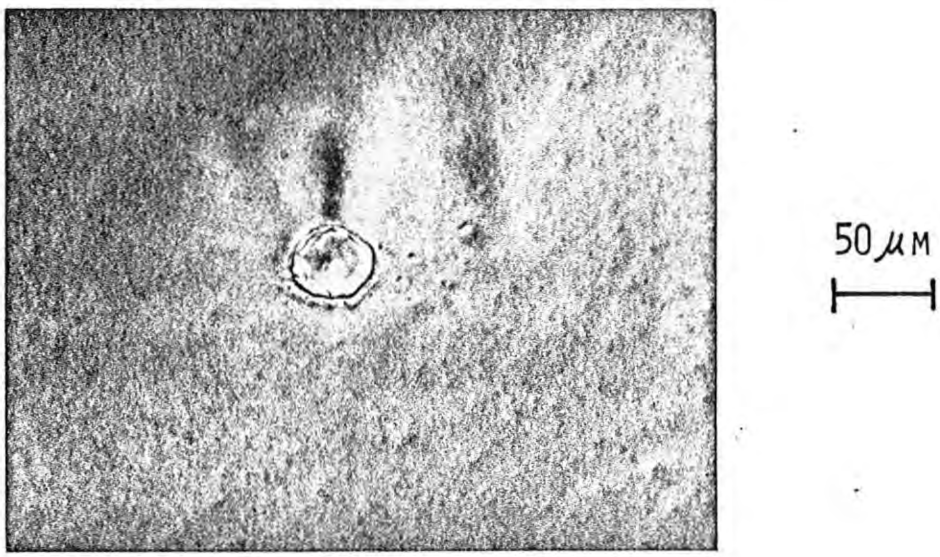
λ/2 As_2S_3 Coating on KCl at 10.6 μm



3 Layer 10.6 μm A-R Coating on KCl

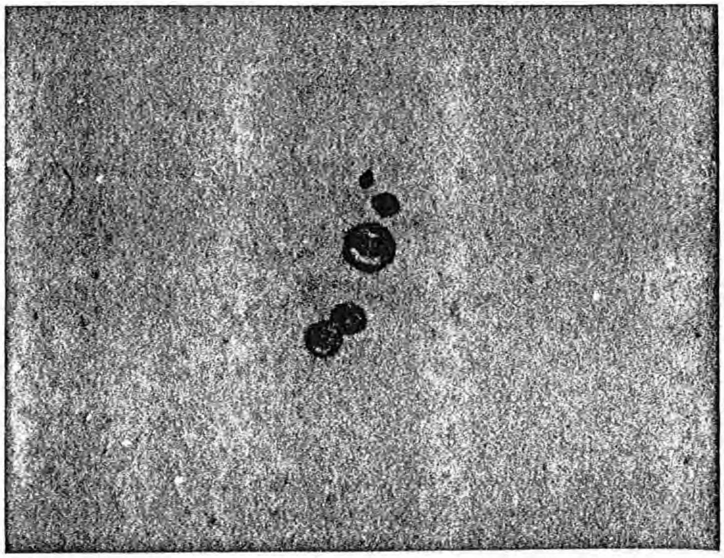


DAMAGE TO $\lambda/2$ - THICKNESS As_2S_3 ON KCL SUBSTRATE

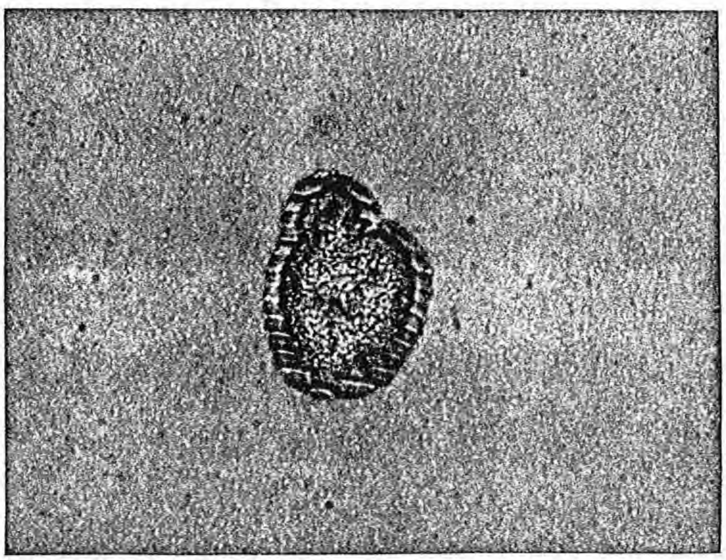


TRANSMITTED PHASE CONTRAST ILLUMINATION

DAMAGE TO $\lambda/2$ THICKNESS As_2Se_3 ON KCL SUBSTRATE



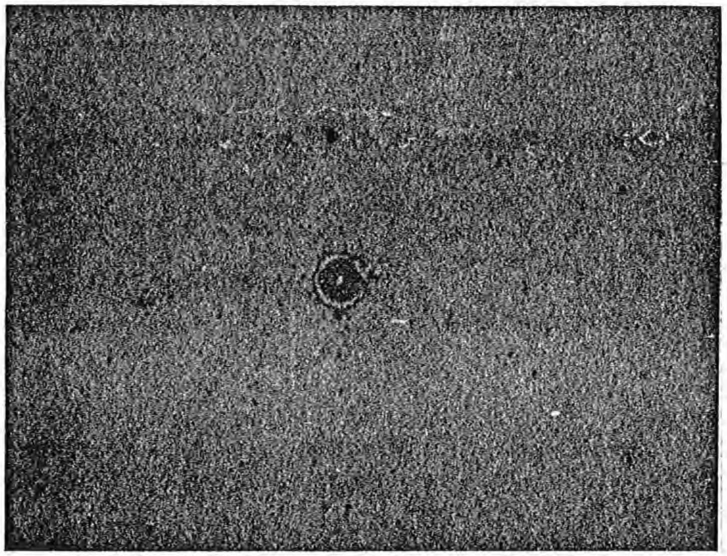
50 μM



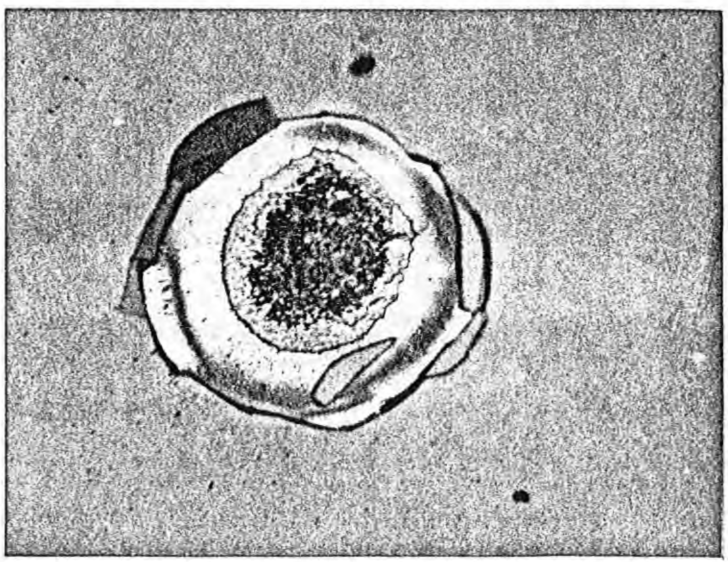
50 μM

REFLECTED NORMAL INCIDENCE ILLUMINATION

DAMAGE TO λ THICKNESS As_2Se_3 ON KCL SUBSTRATE



25 μM

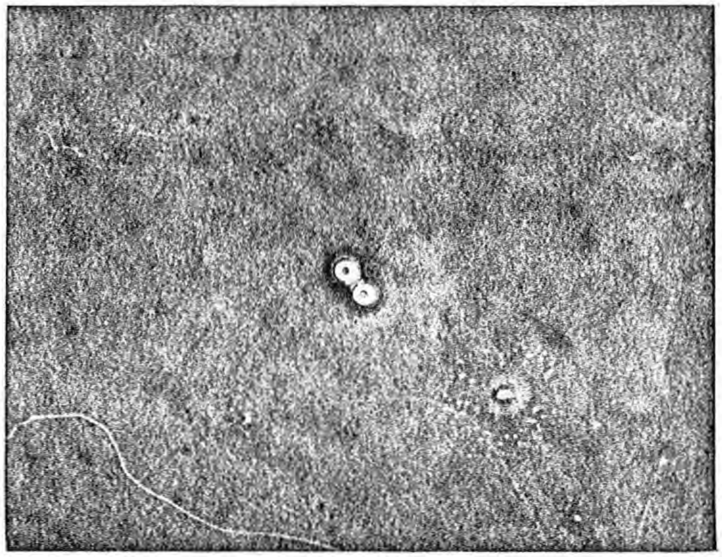


50 μM

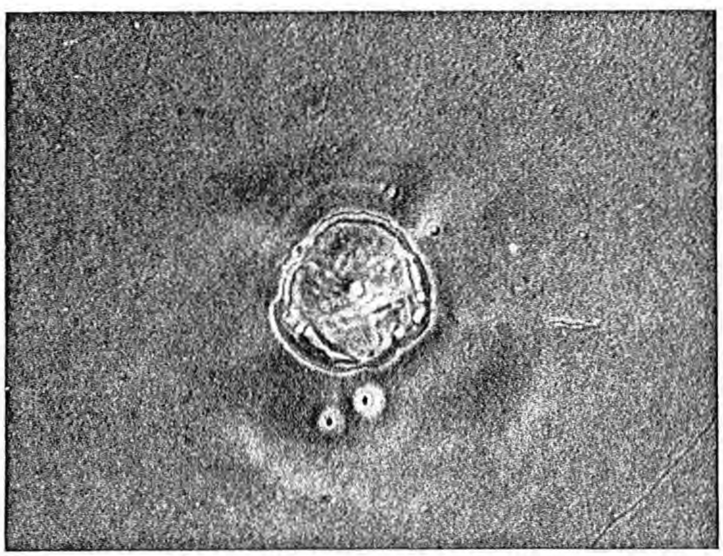
REFLECTED NORMAL INCIDENCE ILLUMINATION

DAMAGE TO $As_2S_3/KCl/As_2S_3$

ON KCl SUBSTRATE



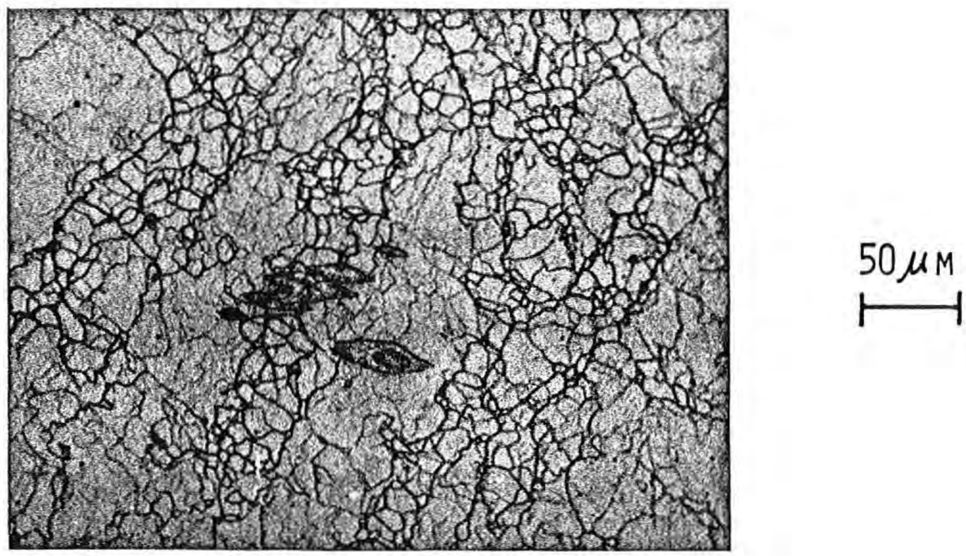
50 μM



50 μM

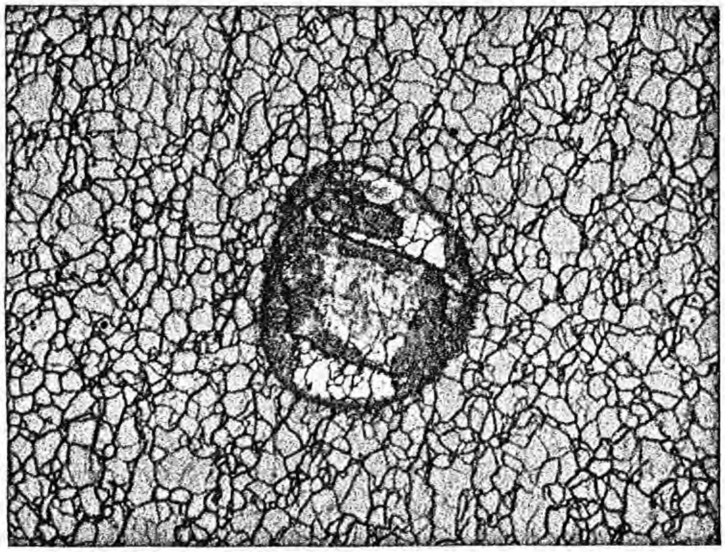
TRANSMITTED PHASE CONTRAST ILLUMINATION

DAMAGE TO $As_2Se_3/KCl/As_2Se_3$
ON POLYCRYSTALINE KCL SUBSTRATE



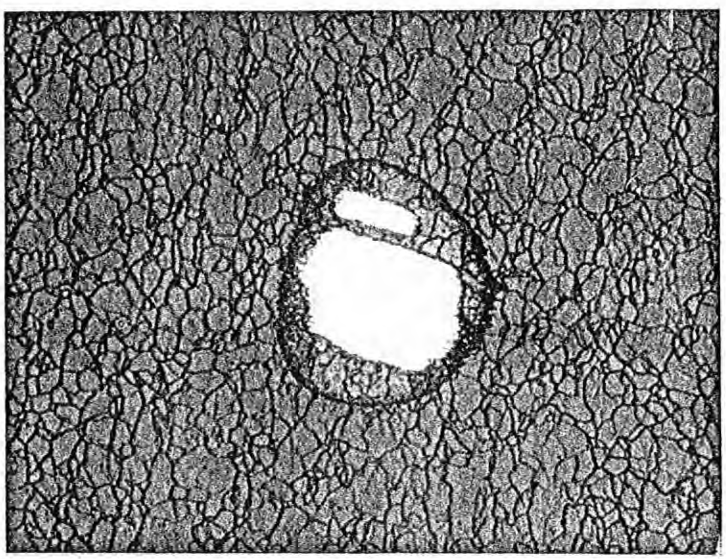
REFLECTED NORMAL INCIDENCE ILLUMINATION

DAMAGE TO $As_2Se_3/KCl/As_2Se_3$
ON POLYCRYSTALINE KCL SUBSTRATE



50 μM

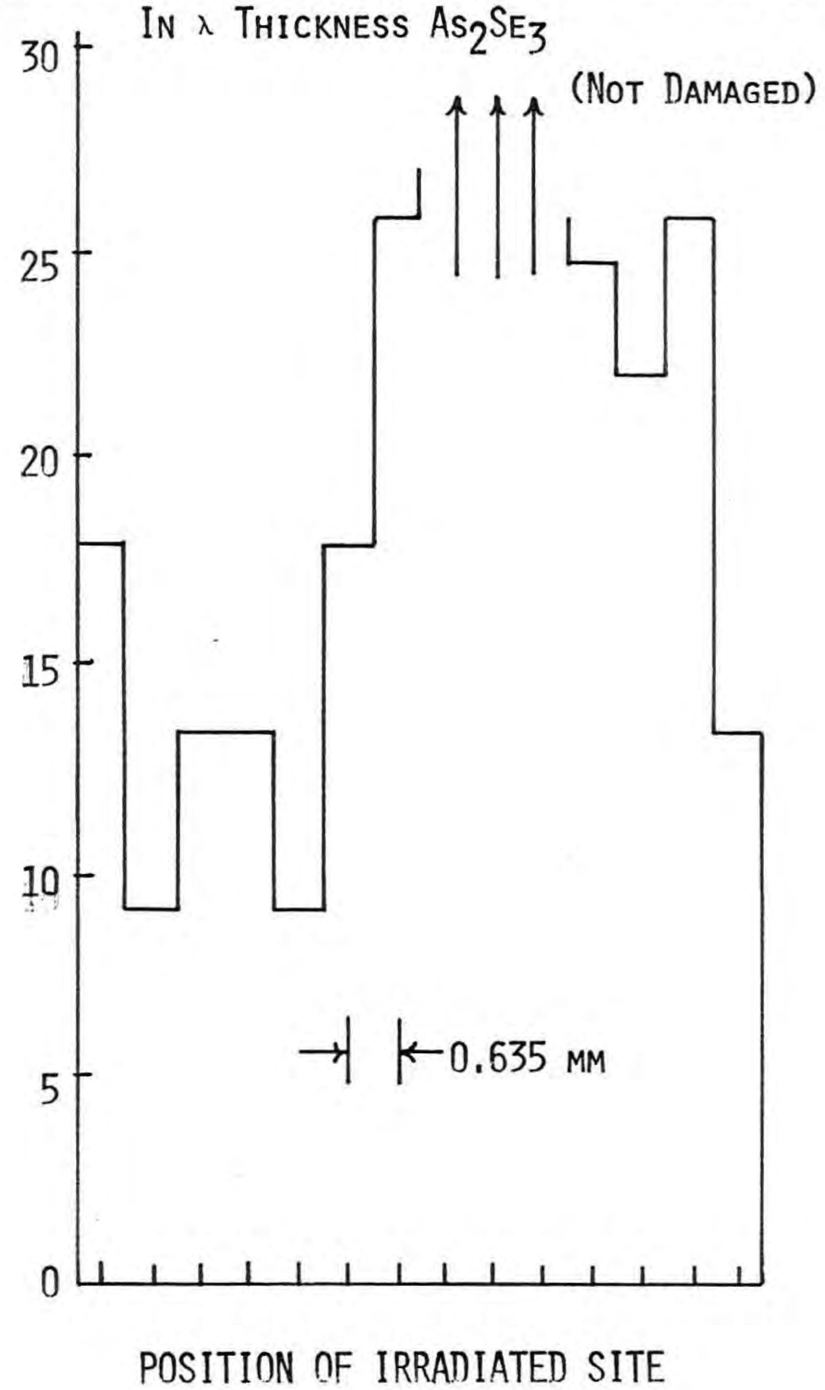
REFLECTED NORMAL INCIDENCE ILLUMINATION



50 μM

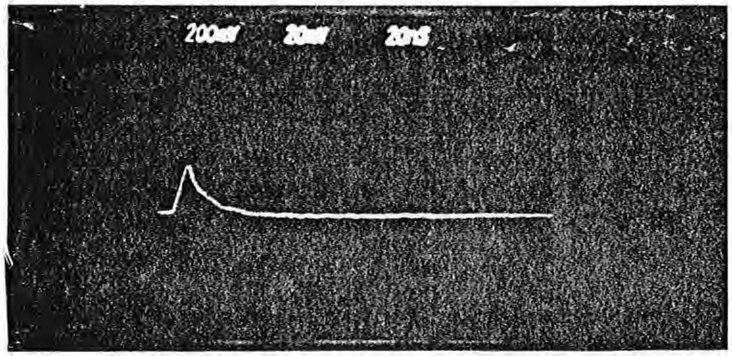
TRANSMITTED BRIGHT FIELD ILLUMINATION

EFFECTS OF COATING NON-UNIFORMITY ON DAMAGE THRESHOLD

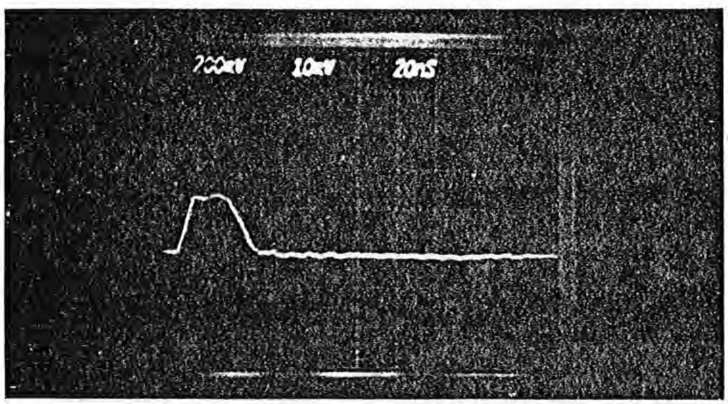


TRANSMITTED WAVEFORM DISTORTION DUE TO 10.6 μM LASER DAMAGE ON A ZnSe/KCl/ZnSe THIN FILM COATING

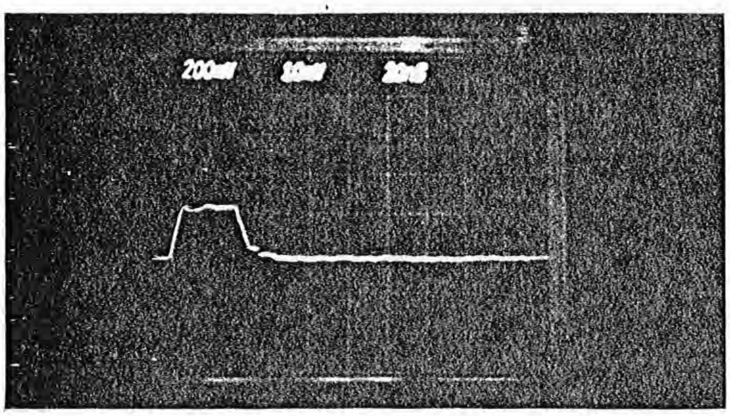
A Flat Topped Pulse Was Used



DAMAGE



DAMAGE



DAMAGE

SURVIVAL CURVE PROOF THAT DAMAGE MECHANISM IS NOT UNIFORM ABSORPTION