SUMMARY REPORT

on

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Laser Modulation of Optical Absorption in ZnSe

(NASA-CR-158789) LASER MODULATION OF OPTICAL ABSORPTION IN ZnSe (Richmond Univ., Va.) 16 p HC A02/MF A01 CSCL 20F

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Submitted to: John Samos
Technology Utilization Officer
NASA-Langley Research Center

By: R. Wayne Major
Department of Physics
University of Richmond
Virginia

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Introduction

Conway has produced LIMA (Laser Induced Modulation of Absorption) at NASA-Langley in ZS and in ZnSe, using relatively high-power laser excitation[1]. Both pulsed and chopped CW beams up to 6 watts have been used. This report deals with the production and study of LIMA in one of the ZnSe crystals used at Langley in earlier work by the author, but carried out in the University of Richmond work with a much lower power laser. The initial question was quite simply whether or not LIMA could be effected and detected at levels induced by such a moderate laser intensity, about 6 mw output. It can be. The modulation is smaller than can be achieved with higher laser photon flux, but is not too difficult to detect. Modulation of the optical absorption on the order of a few parts in 10^5, up to one part in 10^4 appears typical at present. LIMA signals corresponding to a change of one part in 10^3 have been measured by the author at Langley, using a higher power laser.

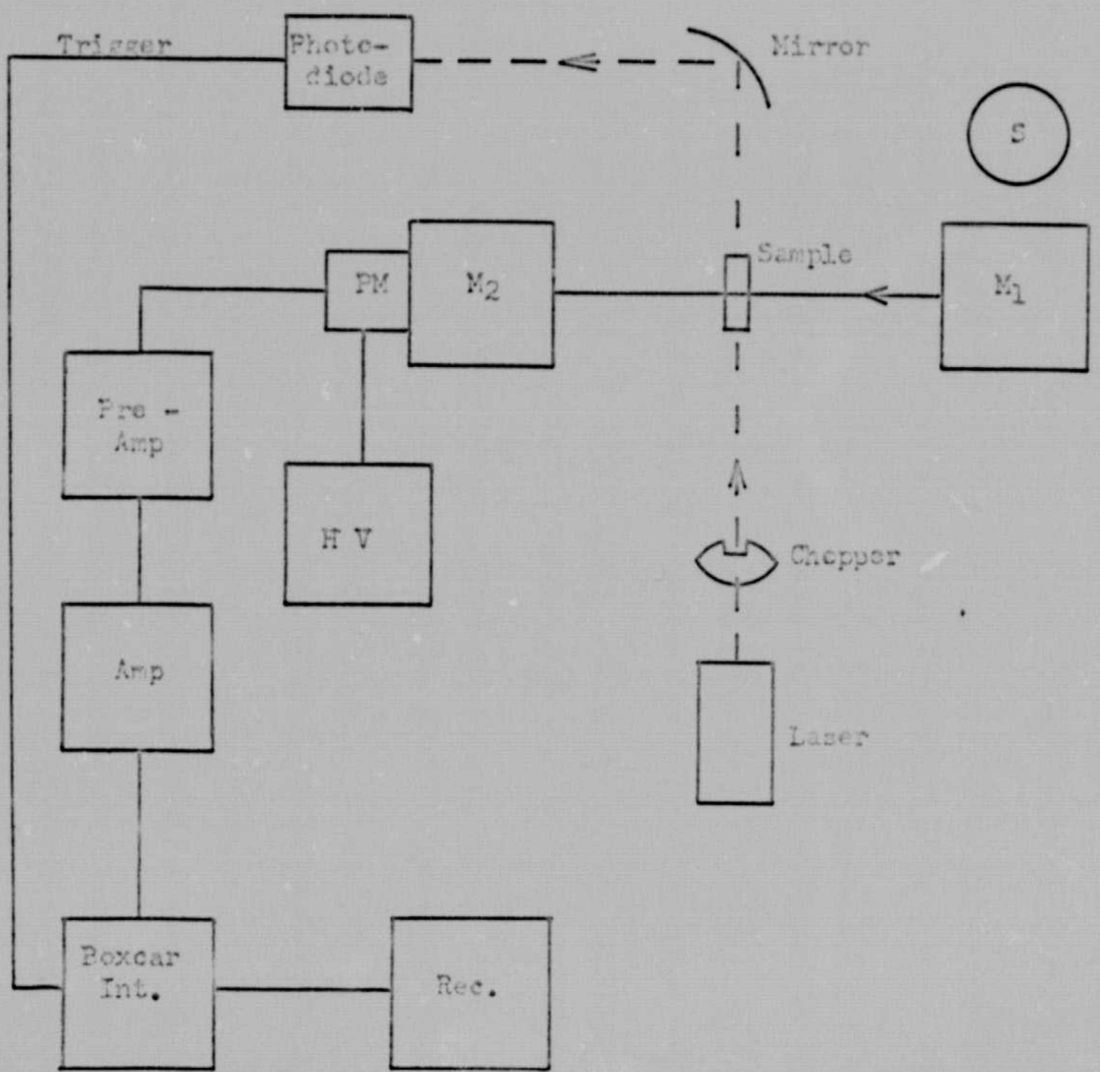
LIMA pulse shapes have been recorded with varying system resolutions at a half dozen wavelengths in the visible, from 4800 Å to 6000 Å. Detailed data have been taken, particularly at 5000 Å, for a range of time intervals following laser pulse turn-on. Quantitative study of the excitation and decay kinetics of these is underway, though analysis is not complete. The following is a presentation of major features, with attention centered upon the detailed measurements made most recently at 5000 Å. Further data are being acquired as this is written.

Experimental System

Both the excitation and detection subsystems were chosen and assembled with economy rather than speed in data acquisition as the governing design factor. The design used (Figure 1) is a modification of that employed by Conway[2], but with the two light beams constrained to be mutually perpendicular in the horizontal plane. The beam of the Spectra-Physics model 120 CW laser (6328 A) is chopped by a homemade 114 Hz. motor-driven disc and enters the sample normal to one of its minimal area faces. Crystal dimensions are approximately ¼" X ¼" X 1/16". The transmitted fraction is focused by a spherical mirror onto a photodiode whose output circuitry produces a square wave voltage pulse 1 to 2 volts high. This synchronously triggers the signal-averager, a P A R model CW-1 boxcar integrator. The signal fed to this integrator originates as a laser-induced change of primary beam intensity seen by an RCA IP-28 photomultiplier. This is operated typically in the range 500 - 700 volts, and looks through a Bausche & Lomb grating monochromator. This small light level change is fed to a low noise, narrow-band preamplifier (P.A.R. CR-4) with a gain of 10^2. Following this is a second stage Furst 220 wide-band amplifier capable of handling the preamplified signal-cum-noise. This yields another gain of 10, for a net gain of 10^3 prior to signal averaging. No significant difference in the final recorded data resulted from trial reversal of these gain factors to 10 and 10^2 respectively.

The adjustable gate of the boxcar advances slowly in real time at a preset rate, sampling successive overlapping portions of a large number of similar pulses, and output is recorded continuously at the scan rate. Random noise is averaged toward zero. A Varian G-11 recorder is used, with 10 mv full scale sensitivity. Signal to noise ratio requirements and detection system noise

Figure 1



Experimental Arrangement

characteristics demand at least several hours' scan time to recover a typical 5 ms signal shape. Commonly used has been a scan time of 6 to 7 hours, corresponding to a total number of pulses on the order of 10^6. While wide gates yield smoother curves more rapidly, good temporal resolution requires as narrow a gate as possible. Gate widths down to 0.21 ms have been used to examine a 5.0 ms duration LIMA signal. The laser rise time is 50 μs; laser pulse duration is 2.5 ms.

While the system is a very inexpensive way to produce and detect LIMA in ZnSe, the quality of observed signals depends upon relatively long-term stability of both the tungsten-filament and laser sources, as well as that of the several detection system components. Replacement of the single-gate instrument by a multi-gate averager such as the P.A.R. model TDH-8 would improve data quality impressively. By thus using all portions of every signal while maintaining present stability, data acquisition times would be reduced by at least an order of magnitude. This in turn would render less significant any long-term drift in either source or detection subsystems.

The experiment design has proved as capable as was expected and originally proposed. Data are reproducible, and there are no extremely critical dimensions or thresholds. Vibration ceased to be a problem when all optical components were mounted on a ground level concrete slab.

Results

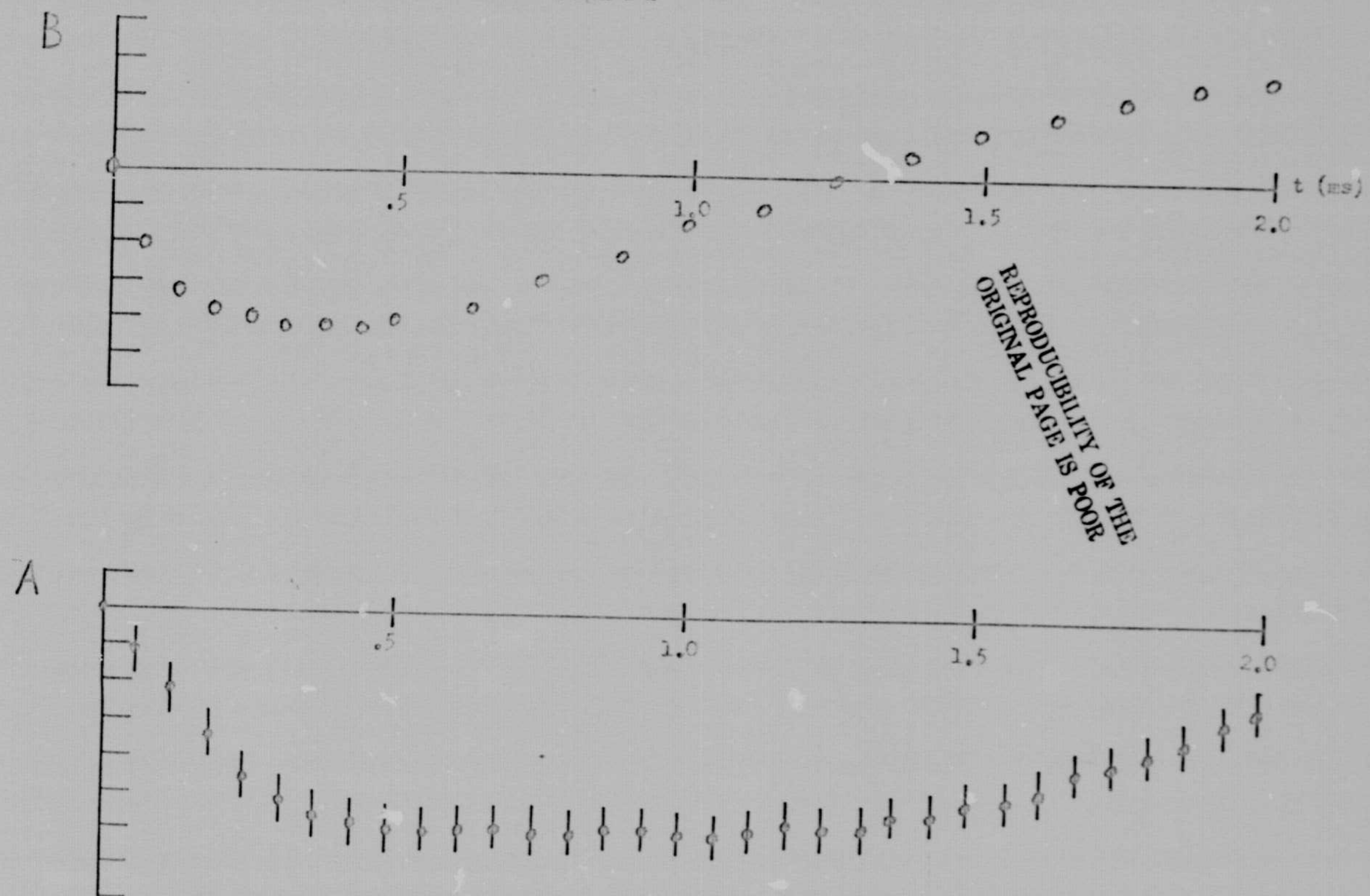
Figures 2 through 5 typify results obtained to date, most of the data therein having been taken since June 1971. LIMA pulse shapes at 5000 Å are displayed for a succession of time resolutions. While LIMA at 5100 Å is similar to that at 5000 Å, the curves are strikingly different at 4800 Å. The major or gross distinction here is that at the latter value, the LIMA pulse appears almost like the 5000 Å result rotated 180° about the time axis.*

Figure 2 shows the result of LIMA pulse shape dependence upon detection system bandwidth during the first two milliseconds of laser irradiation. In each plot shown, the source monochromator slits were set at 0.20 mm. Curve A shows the result for a detection slit of 0.40 mm, while curve B represents superior resolution, corresponding to a 0.20 mm detection slit. All other experimental parameters were identical. Clearly the greater bandwidth of the detector in case A admits appreciable components of LIMA at neighboring wavelengths. Data (not shown) at an intermediate bandwidth (0.30 mm) shows a shape consistent with this statement. While LIMA could be detected at slit openings as small as 0.10 mm, all following data were taken with the slits of both monochromators at 0.20 mm.

Development of the 5000 Å signal in time is extended in Figure 3, and includes an increased - absorption component larger than the initial decreased absorption. The onset of decay of the excitation following laser cut-off at 2.5 ms is evident here. Figure 4 shows a different but quite similar result, recorded in two parts. The decay is obtained by using the negative slope of the laser-synchronous boxcar trigger. It is worth noting that contiguous running times of about 7 hours <u>each</u> were required to record the two 5 ms components shown. This curve shows a feature common to most signals at wavelengths

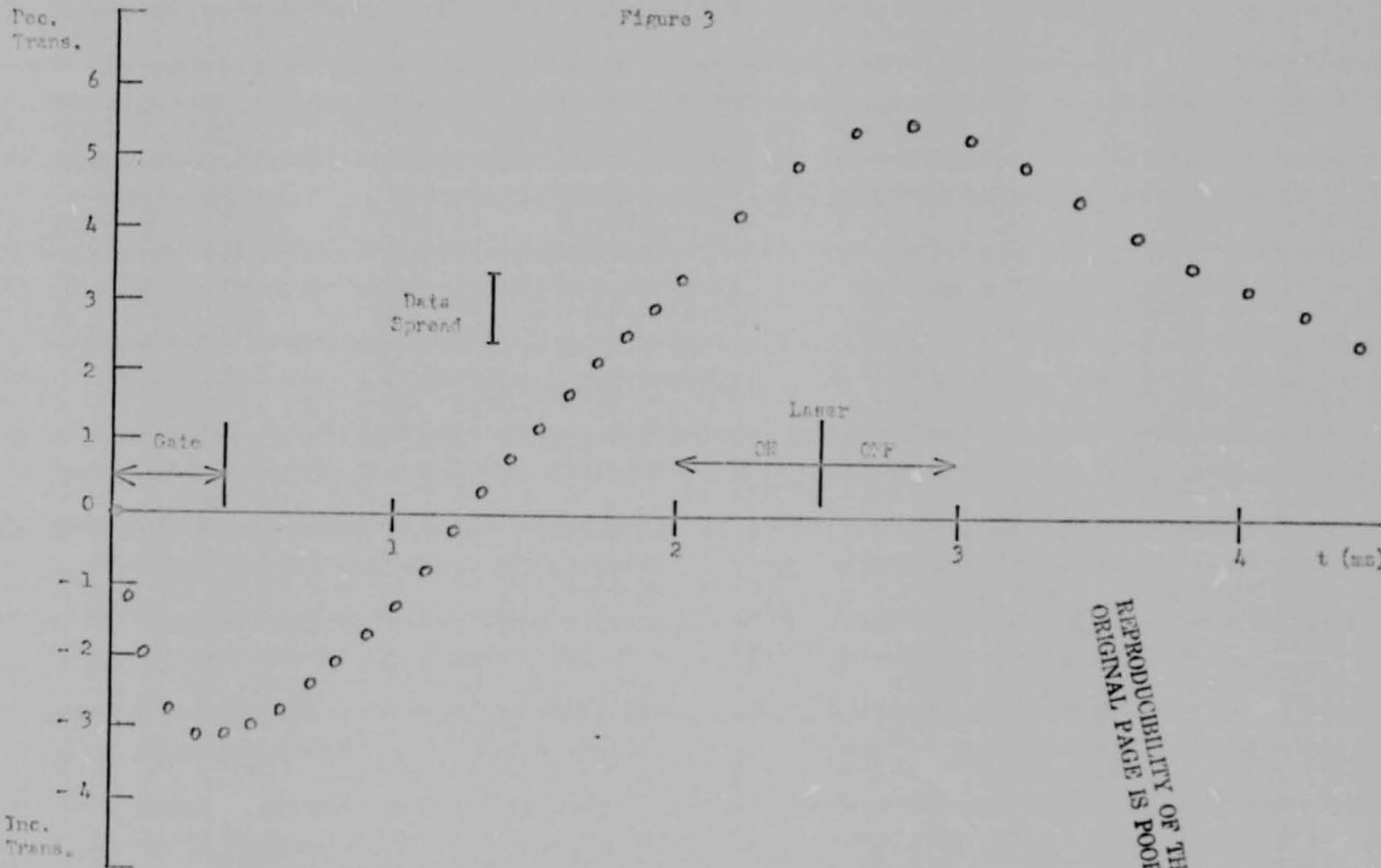
*(Spectral LIMA data taken for the same sample in 1969 (Aug. 6) with a phase-comparison lock-in amplifier (P.A.R. HR-8) showed a near discontinuity in the phase at 4800-4850 Å.)

Figure 2



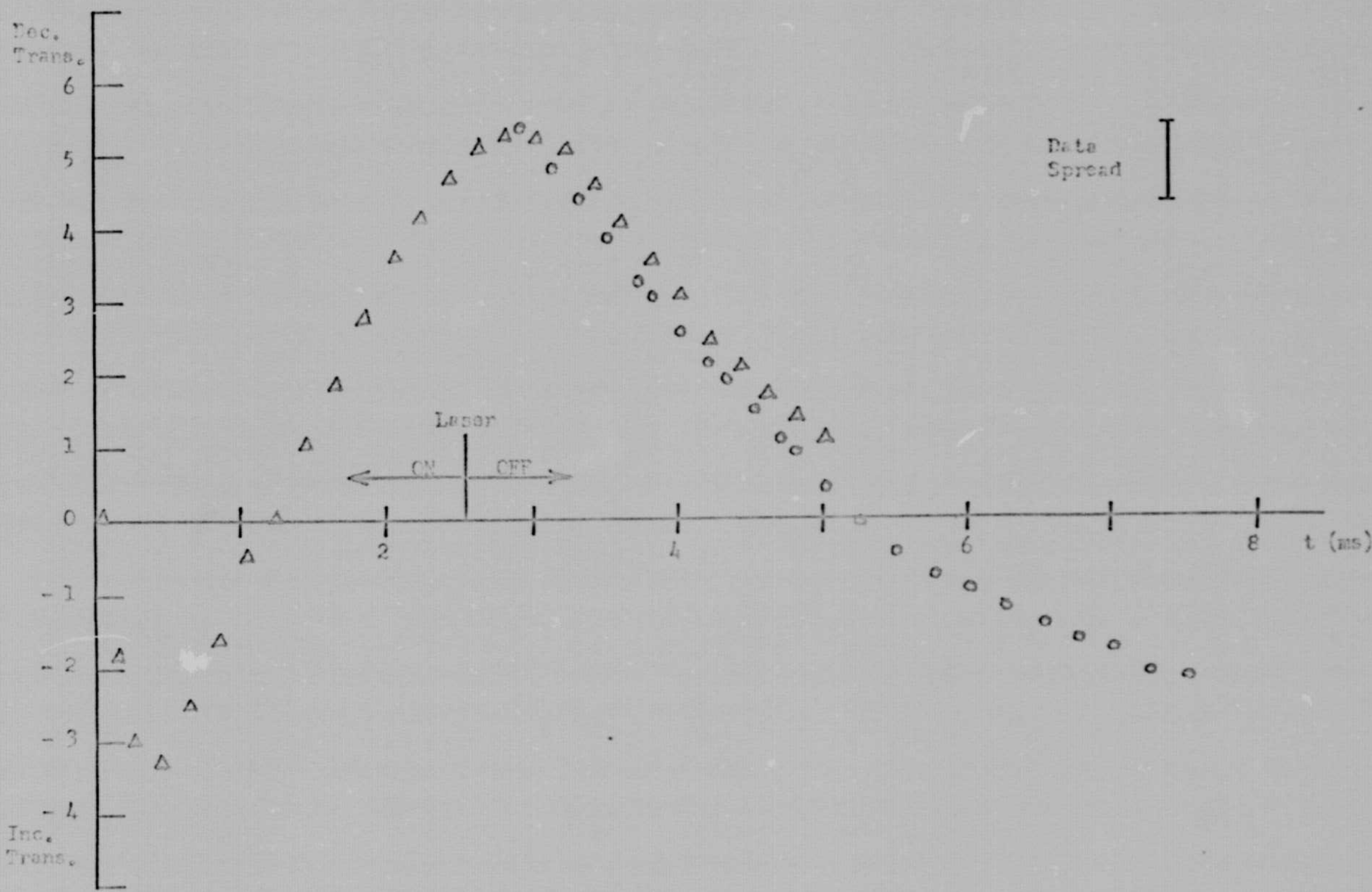
Effect of reducing detection monochromator slit S ; $S_B = S_A/2$.

Figure 3



LIDA at 5000 Å as recorded in arbitrary units (full scale)

Figure 4



LIMA at 5000 Å. Overlapping 5 - ms runs recorded consecutively. Full scale in arbitrary units.

used to date: final decay towards the original polarity. In this case, it is clear that within 2.5 ms after laser cut-off, the modulation has decayed to nearly the same level of decreased absorption as appears in the first 0.4 ms. It is tempting to associate the initial maximum with the lingering tail of this decay, and to assume that in fact the presence of the initial maximum is due to the failure of the crystal to return to a completely relaxed (unexcited) state prior to the onset of the next laser pulse. The 'dual-polarity' character is in qualitative agreement with earlier work done by the author at Langley.

Figure 5 demonstrates another consistency in the 5000Å data. The solid curve shows the onset of increased absorption after the first millisecond of laser illumination. The large flagged triangles are taken from an enlargement of a polaroid record of LIMA in the same sample, made by the author in 1969 at Langley. Fewer data points are available, and the temporal resolution is poor on the millisecond scale, but qualitative agreement is evident. The latter data were produced by a slowly chopped (13 Hz) 6 watt cw laser of 5682 Å wavelength. The five points shown represent only 5% of the complete (40 ms) LIMA signal recorded, and the laser pulse exhibited a rise time of about 1 ms. In contrast, the laser rise time is 50 μs for the present data (solid curve). Conway has shown that, in CdS at least, LIMA spectra are not shape-sensitive to laser intensity.[3] The result suggests that the <u>rate</u> of the excitation responsible for LIMA in this case is also independent of laser power. For example, the higher power laser pulse almost certainly reached more than 6 mw within the first 50 μs of its (slower) rise.

It should be noted that the data recorded <u>during</u> an initial time interval equal to one integrator gatewidth is of little significance, since the instrument requires a full gatewidth to respond to a step function. The behavior of all the signals observed in the first several milliseconds, however, is the same. All data at a given wavelength show the 'polarity' reversal, or crossover occurring at the same time.

Figure 5

In the case of the 5000 Å data shown (e.g. Fig. 3) this occurs 1.2 ms after the onset of laser excitation, or nearly halfway through the 2.5 ms laser pulse.

Figure 6 is a semilog treatment of the decay of the same LIMA signal shown in Fig. 4. Though evidently not quite complete, the decay appears simply exponential with but one rate. Curve 6-A represents the choice of the final data point as the decay limit, or zero. More realistically, curve 6-B is based upon a lowering (by 7% of maximum signal amplitude) of this limit, and the result is more nearly linear. From either curve, the time constant for the decay is 1.9±.1 ms. No distinct 'fast' or 'slow' components are distinguishable in these data. Measurement of the decay is presently underway for longer time intervals. Checks have been made to ensure that luminescence due to laser illumination only was not observable.

The apparent simple nature of the decay as suggested above is not readily reconciled with the fact that LIMA also shows, at nearly all wavelengths, a rapid establishment at one polarity followed by the slower development of a dominant contribution of the opposite sense. The latter continues to develop in magnitude throughout the laser pulse duration. The initial relative maximum could be in fact a near discontinuity in time, to within the one-gatewidth resolution limit of the boxcar for such an input.

Figure 7 shows a sketch (dashed) of a typical LIMA signal as discussed in this report, with a hypothetical dismemberment into two components of opposite polarity (solid curves). Observed decay could be phenomenologically interpreted as consisting of the sum of these. The lower, increased-transmission component is assumed to be of thermal origin. In fact there is some basis for such an assumption. An increase of approximately 1-2% in the dc transmission of the monochromatic primary beam has been observed many times when the laser is first turned on before a data run. The dc volt meter used to record light level at the

Figure 6

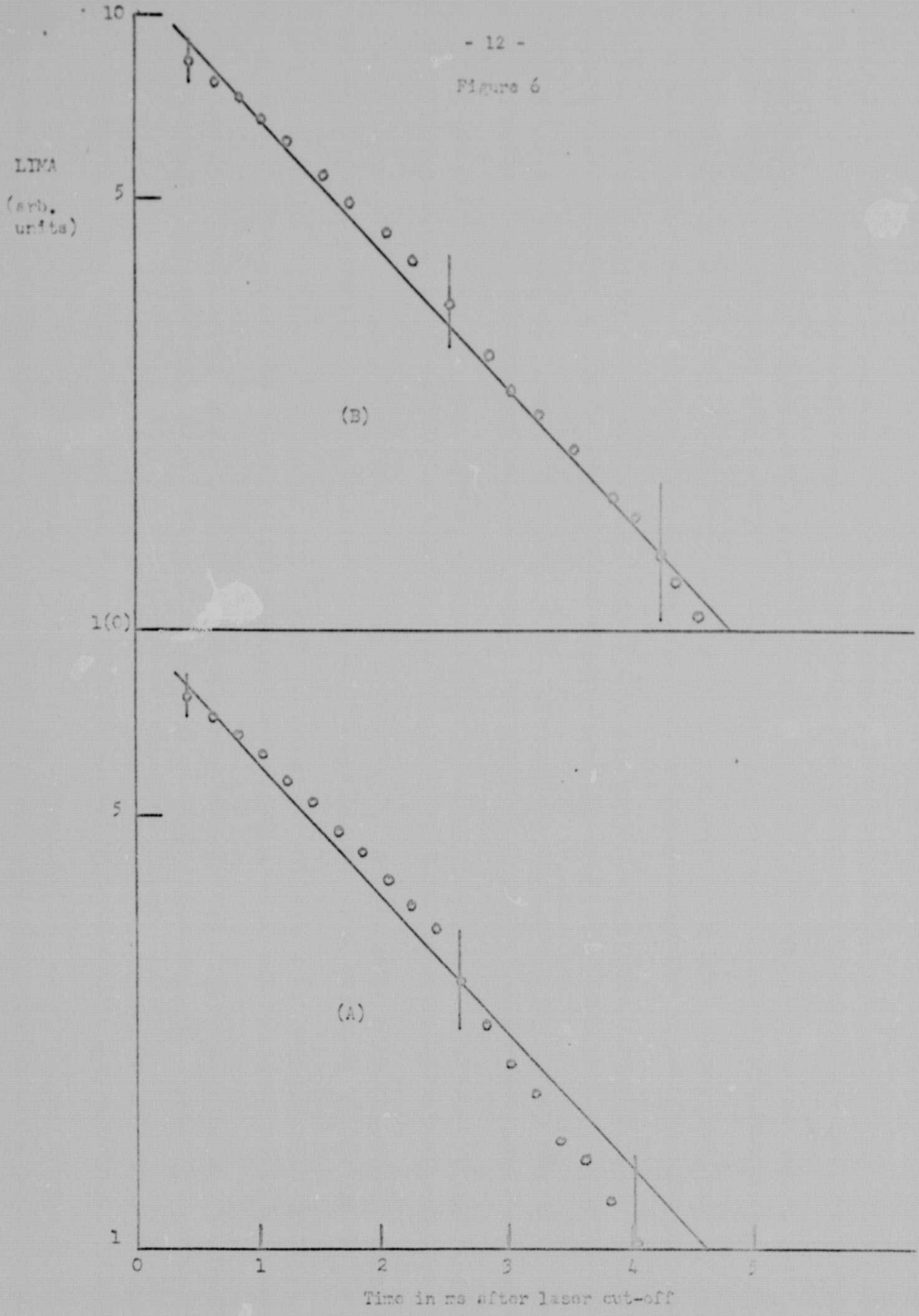
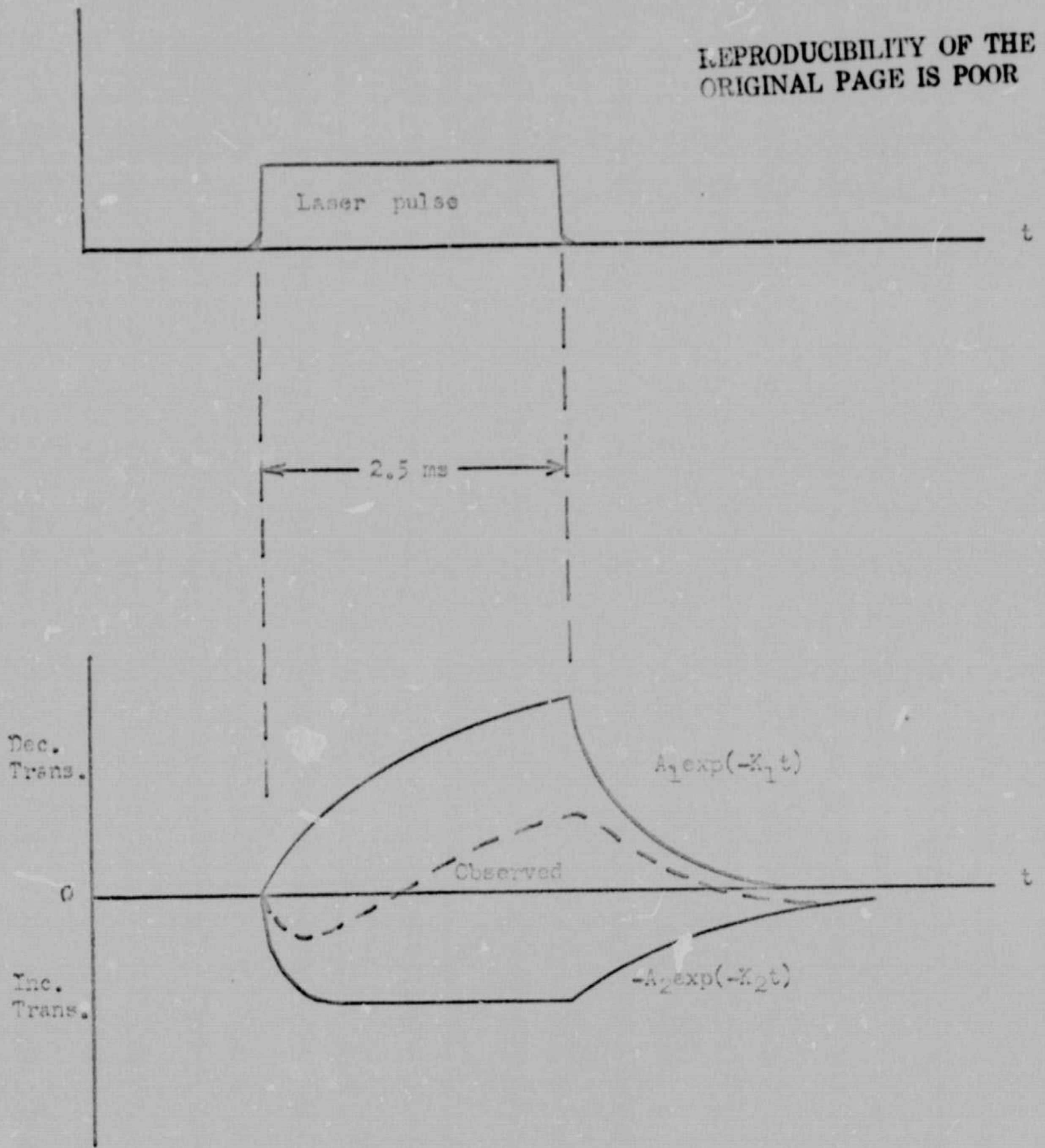

LIMA
(arb. units)
10
5
1(0)
5
1
(B)
(A)
0
1
2
3
4
5
Time in ms after laser cut-off

Figure 7


Laser pulse
t
2.5 ms
Dec. Trans.
$A_1\exp(-K_1t)$
Observed
0
t
Inc. Trans.
$-A_2\exp(-K_2t)$

P-M output shows an increase corresponding to increased transmission and requiring 5 to 10 seconds to achieve a steady state. Certainly the crystal has warmed from its initial ambient temperature. What is required to fit the above tentative schematic is that superimposed on this time-average dc increase is a small "ripple" in the temperature at the laser frequency. Such a temperature ripple could provide the increased transmission component suggested, via transient effects on the phonon spectrum. Whether or not the accompanying development of a decreased transmission component (upper curve) could be independent of this thermally-induced or phonon component is not clear. Note that such a kinetic model requires that the former decay faster than the latter (i.e. $K_1 > K_2$) and from a greater signal amplitude. Finally, while the observed decay tail demands the inequality, the two rates cannot differ greatly if the overall result is to be nearly exponential, as in Fig. 6. That is, if $y_1 = A_1 \exp(-k_1 t)$ and $y_2 = -A_2 \exp(-k_2 t)$, then

$$y_{net} = y_1 + y_2 = (A_1 - A_2) - (A_1 K_1 - A_2 c K_1) t + (A_1 K_1^2 - A_2 c^2 K_1^2) t^2/2 - \cdots, \quad c K_1 = K_2 .$$

Observing that this is truly exponential only if $c = 1$, then for values of c not too different from unity one might satisfy the required inequality while retaining a near exponential sum as observed in Fig. 6.

If a single trapping level dominates the decay or its (hypothetical) decreased transmission component as outlined above, and if the trap concentration exceeds that of carriers by several orders of magnitude, then one expects a simple exponential with time constant $T = (NA)^{-1}$. N is the trap concentration, A a recombination coefficient.[4] This simple model neglects thermal ionization of trapped carriers, so actual carrier lifetime may be considerably less than the 1.9 ms characteristic of the data.

References

1. K. W. Boer and E. J. Conway; Light Induced Modulation of Absorption in CdS. NASA T. N. D-4466, March, 1968.

2. E. J. Conway; Light Induced Modulation of Broad Band Optical Absorption in CdS. J. Appl. Phys. 41, No. 4, 1689, 1970.

3. E. J. Conway; Light Induced Modulation of Absorption and Photoconductance of Single Crystal CdS. Paper presented at Nov. 1968 A.P.S. meeting. (Ref. pg. 4 of reprint).

4. C. Kittell; Solid State Physics, J. Wiley & Sons. 1967 (3rd Ed.) pg. 551.