LBL-12695
Preprint

Lawrence Berkeley Laboratory
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Materials & Molecular
Research Division

Submitted to IEEE Journal of Quantum Electronics

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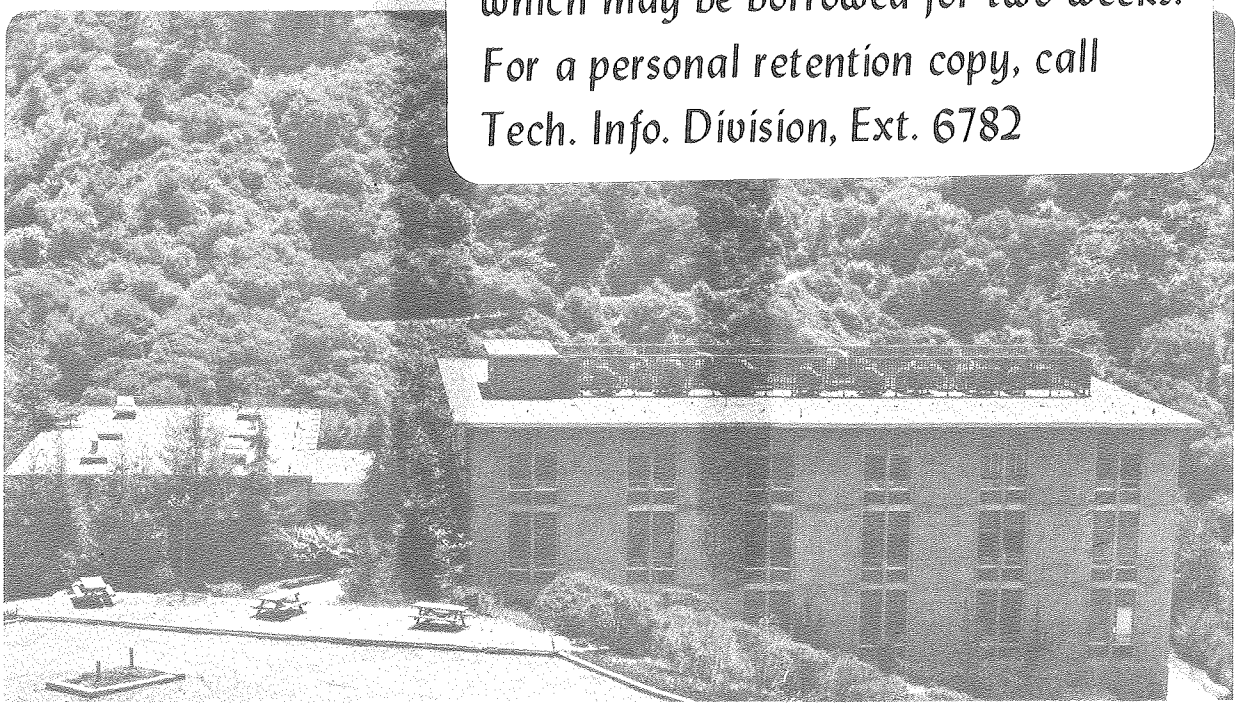
May 1981



Prepared for the U.S. Department of Energy under Contract W-7405-ENG-48

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PICOSECOND PULSE SHORTENING USING DYE #5 AS A SATURABLE ABSORBER

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Abstract

The effects of the new infrared absorbing dye #5 used as a saturable absorber are studied. Picosecond light pulse width and mean squared intensity reductions are measured employing two photon fluorescence. The saturable absorber effects of dye #5 are compared to dye #9860 at O.D. = 2.0. The results agree qualitatively with theoretical predictions for both the pulse shortening ratio's and the pulse intensity transmission's dependence on the saturable absorber optical density.

1. Introduction

The shortest possible light pulses are desired for optimum time resolution in picosecond studies. One method used to shorten a light pulse is to pass the pulse through a saturable absorber external to the laser cavity [1,2]. Although some theory for a saturable absorber's effect on a picosecond light pulse has been presented [2], there has been little experimental work on the pulse shortening or intensity reducing effects of external saturable absorbers.

Recently, Reynolds and Drexhage [3], reported on a new infrared absorbing dye, Kodak heptamethine pyrylium dye #5, which has a very fast absorption recovery time of 2.7 psec compared to 6.5 psec for Kodak dye #9860 [4]. Others have reported [4,5] that dye #5 used as a mode-locking dye in Nd:glass laser oscillators gave pulses substantially shorter than those obtained using dye #9860. Kopainsky et al. [4] have also suggested the use of dye #5 as an external saturable absorber. We report on the effect of dye #5 used as an external saturable absorber on the width and intensity of single, high power pulses from a Nd:glass oscillator/amplifier system. The experimental results are compared to the results of a theoretical model. In addition, the saturable absorber effects of dye #5 and dye #9860 are compared at optical density (O.D.) = 2.0.

2. Experimental

A single picosecond pulse was selected from the rising edge of pulse trains emitted by a TEM_{00} selective, passively mode-locked Nd:glass laser oscillator [6]. A Pockels cell driven by a low jitter electronic circuit [7] consistently selected the 6^{th}- 8^{th} intense pulse from pulse trains of 30-45 pulses. A fast photodiode and a Tektronix 7834 fast storage oscilloscope monitored the pulse trains. Shots which displayed multiple pulse trains were excluded from the data.

The selected pulse was amplified in a three stage Nd:glass amplifier. The pulse energy after amplification was determined to be approximately 30 mJ using a Korad/Hadron Model 100 thermopile. Throughout this study, the pulse energy was kept constant.

Dye #5 and dye #9860 were obtained from Eastman Kodak. The dyes were dissolved in 1,2-dichloroethane and the optical densities were measured at 1.06 μm in a 1 cm cell. The dye solution was placed in a 1 cm cell after the final amplifier. The laser pulse burn spots on developed Polaroid film positioned in front of the dye cell had a diameter of 5-6 mm and showed no substructure.

To measure pulse widths, the entire unfocused pulse was sent into a standard two photon fluorescence (TPF) apparatus [8]. The TPF signal was obtained from a

1×10^{-3} M methanol solution of rhodamine 6G in a 1 cm cell. An 85 mm camera lens imaged the TPF trace with a magnification of approximately 1.0 on a Princeton Applied Research Model 1254 optical multichannel analyzer (OMA) ISIT detector. High f-numbers of f/22 to f/8 were used to maximize the depth of field. The OMA was interfaced to a minicomputer which displayed the TPF intensity plot on a CRT screen. Calibration, focus and resolution of the lens-OMA system were determined using a calibrated fine copper mesh at the TPF cell position.

3. Results

Several TPF screen displays are shown in Figure 1. The display is linear with respect to TPF intensity. TPF peak-to-background ratios were in the range 2.8 - 3.0 to 1 on all shots. TPF intensity traces taken without a saturable absorber are shown in Figures 1a and 1b. Figures 1c and 1d show TPF intensity traces taken using O.D. = 2.0 saturable absorbers dye #9860 and dye #5, respectively.

Pulse widths were determined by measuring the full width at half maximum of the TPF intensity traces. In all figures, each point is an average of 20-30 measurements, and the bars show the standard deviation of the data for each point. Figure 2 summarizes the TPF autocorrelation widths measured with no saturable absorber and with dye #5 at O.D. = 1.0, 2.0 and 3.0. All data were taken on the same day under identical amplification and pulse selection conditions.

Assuming Gaussian pulses, the pulse widths, t_p, were calculated using the formula $t_p = 2\,\Delta z n/(2)^{1/2}c$, where Δz is the measured TPF autocorrelation width, n is the refractive index at 1.06 μm in the TPF cell, and c is the speed of light [9,10]. Measured methanol refractive indices in the visible were extrapolated to give $n \simeq 1.3235$ at 1.06 μm. The measured pulse shortening ratios were 0.84, 0.74 and 0.71 at optical densities 1.0, 2.0 and 3.0, respectively.

The light pulse's mean squared intensity after passing through the saturable absorber was determined from the peak TPF intensity [8]. Changes in the mean squared intensity emphasize intensity changes near the peak of the pulse. Figure 3 shows the relative TPF peak intensities measured with no saturable absorber and with dye #5 at O.D. = 1.0, 2.0 and 3.0.

Dye #5 and dye #9860 were compared by measuring TPF autocorrelation widths and TPF peak intensities with both dyes at O.D. = 2.0. Measurements were made on the same day at constant amplification and selected pulse energy. The results are shown in Figures 4 and 5. Dye #5 gives a shorter TPF autocorrelation width and a greater reduction in pulse intensity than dye #9860 at the same optical density.

Davis and Lin [11] have suggested that optical nutation effects can cause the break up of high intensity pulses traveling through a saturable absorber. However, the TPF traces observed in this study were almost always smooth and showed no evidence of a coherence spike, which would be associated with pulse substructure [8]. Therefore, pulse break up does not occur under the conditions of this experiment.

4. Discussion

Penzkofer [2] has numerically investigated light pulse shortening and intensity reduction effects using saturable absorber parameters modeled for dye #9860. His calculations suggest that dyes having a smaller relaxation time to pulse width ratio should show qualitatively similar behavior (see fig. 5 of ref. 2).

Penzkofer presented a graphical summary of the numerical results for the pulse shortening ratio (fig. 4 of ref. 2) and for the peak intensity transmission (fig. 8 of ref. 2) as a function of input peak intensity and saturable absorber optical density. Both the pulse shortening ratio and the peak intensity transmission show qualitatively different behavior at low input peak intensities of $1 - 6 \times 10^8$ W cm^{-2} compared to the behavior with input peak intensities above 6×10^8 W cm^{-2}.

At low input peak intensities, the calculated pulse shortening ratios decrease with increasing optical density, then level off, with ratios remaining in the range 0.9 - 0.5 for optical densities greater than O.D. = 1 - 5. In the same low intensity region, the peak intensity transmissions show a steady decrease with increasing optical density.

With input peak intensities above 6×10^8 W cm^{-2}, the pulse shortening ratios do not level off until much higher optical densities of 7 - 40, at which point very

small pulse shortening ratios of 0.5 - 0.2 are obtained. With high intensity pulses, the peak intensity transmissions do not decrease significantly at low optical densities, but show a sudden reduction above a particular transition optical density in the range O.D. = 5 - 40.

Our experimental pulse shortening ratios begin to level off at a value of 0.7 for O.D. = 3.0, while the measured mean squared intensity transmissions show a steady decrease with increasing optical density. These results are qualitatively consistent with Penzkofer's numerical results in the low input peak intensity range 1 - 6 x 10^8 W cm^{-2}. Because the different relaxation time and cross section of dye #5 relative to dye #9860 may affect the input intensity level at which dye saturation effects are observed, a quantitative comparison with Penzkofer's results is not possible. We note that the pulses used in this study had peak intensities of several GW cm^{-2}, indicating that dye #5 at high input peak intensities gives saturable absorber effects qualitatively similar to dye #9860 effects expected at lower input peak intensities.

In conclusion, dye #5 has been shown to be effective in shortening high power Nd:glass laser pulses, but at the cost of significant intensity reductions. Dye #5's short relaxation time enables this saturable absorber to be more

effective in pulse shortening than dye #9860 at the same optical density. The behavior of dye #5 is qualitatively similar to the behavior predicted by a numerical model.

Acknowledgements

This work was supported by the Director, Office of Energy Research, Office of Basic Energy Sciences, Chemical Sciences Division of the U.S. Department of Energy under Contract Number W-7405-ENG-48. ALH and MB gratefully acknowledge support from the National Science Foundation for graduate fellowships. The authors would like to thank Steven J. Davis for his design and help in the construction of the low jitter electronic pulse selector. We are also appreciative to Roman Hawryluk of Eastman Kodak for providing Kodak dye #5.

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Figure Captions

Fig. 1 CRT screen traces of TPF intensity profiles measured by an OMA for constant energy light pulses using (a) and (b) no saturable absorber, (c) saturable absorber dye #9860 at O.D. = 2.0 and (d) saturable absorber dye #5 at O.D. = 2.0.

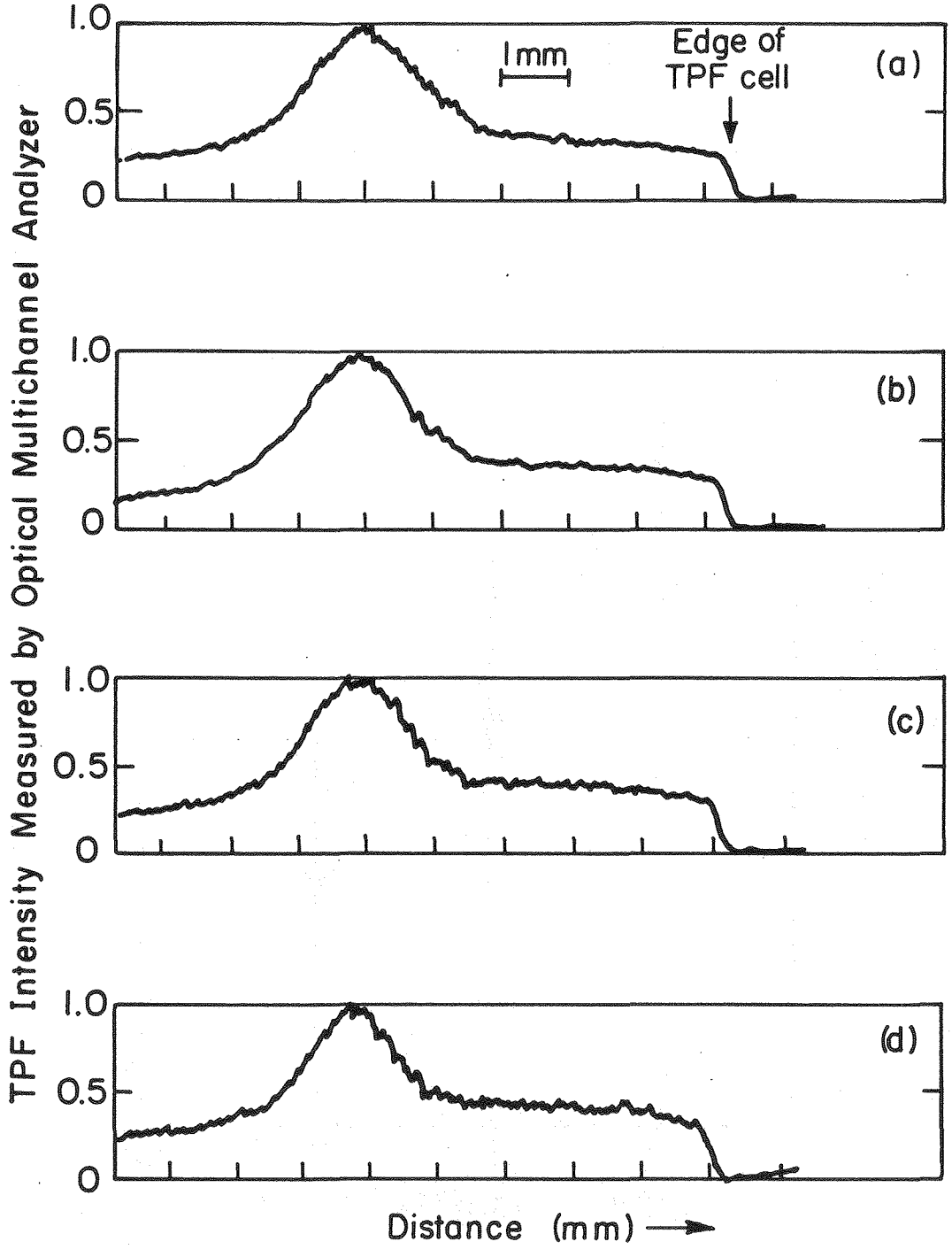
Fig. 2 Pulse autocorrelation width dependence on optical density of saturable absorber dye #5 using constant energy light pulses.

Fig. 3 TPF peak intensity dependence on optical density of saturable absorber dye #5 using constant energy light pulses.

Fig. 4 Same day comparison of pulse autocorrelation width dependence on saturable absorber dye #9860 and dye #5 at O.D. = 2.0 using constant energy light pulses.

Fig. 5 Same day comparison of TPF peak intensity dependence on saturable absorber dye #9860 and dye #5 at O.D. = 2.0 using constant energy light pulses.

Figure 1


1.0
0.5
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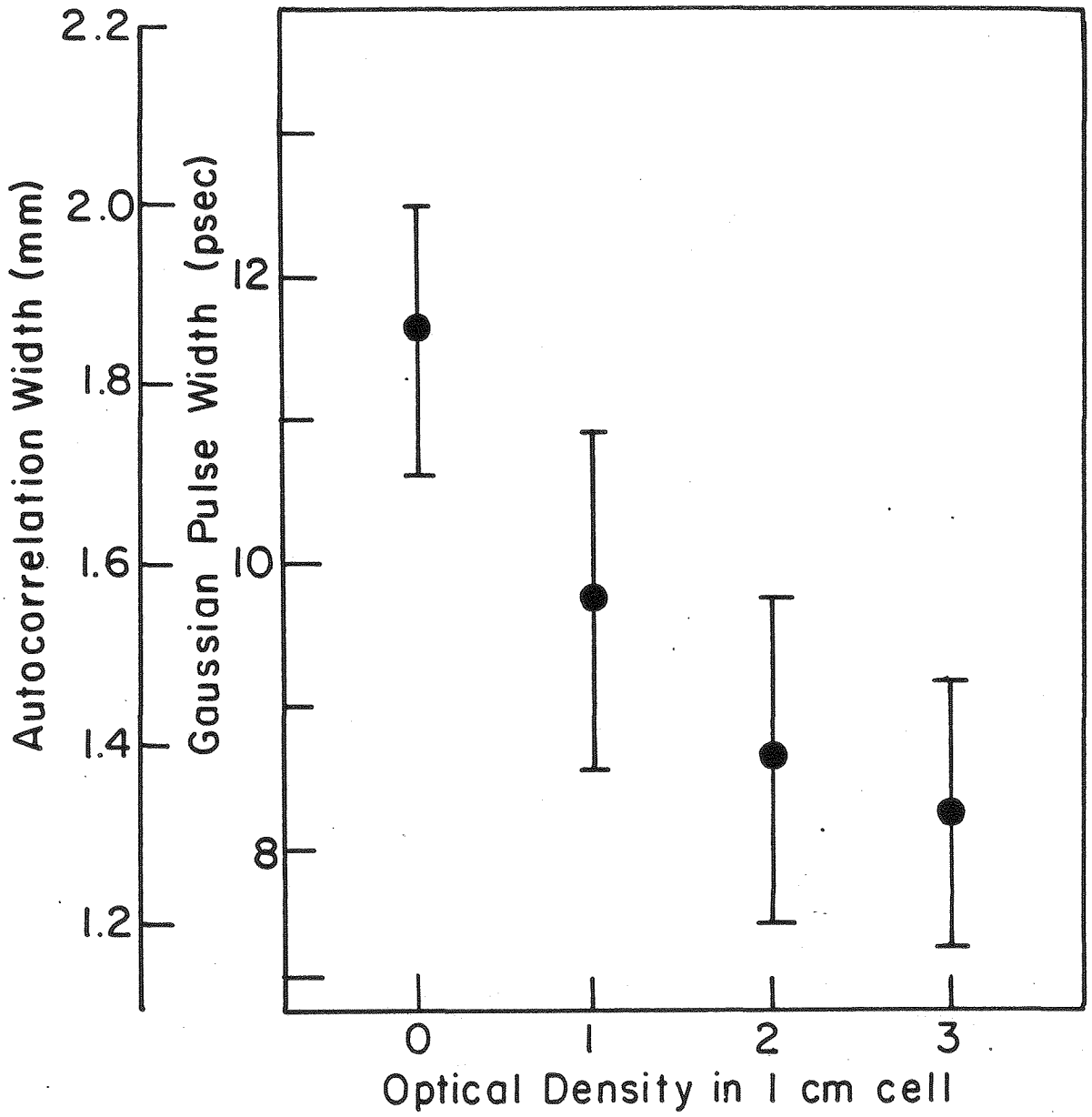
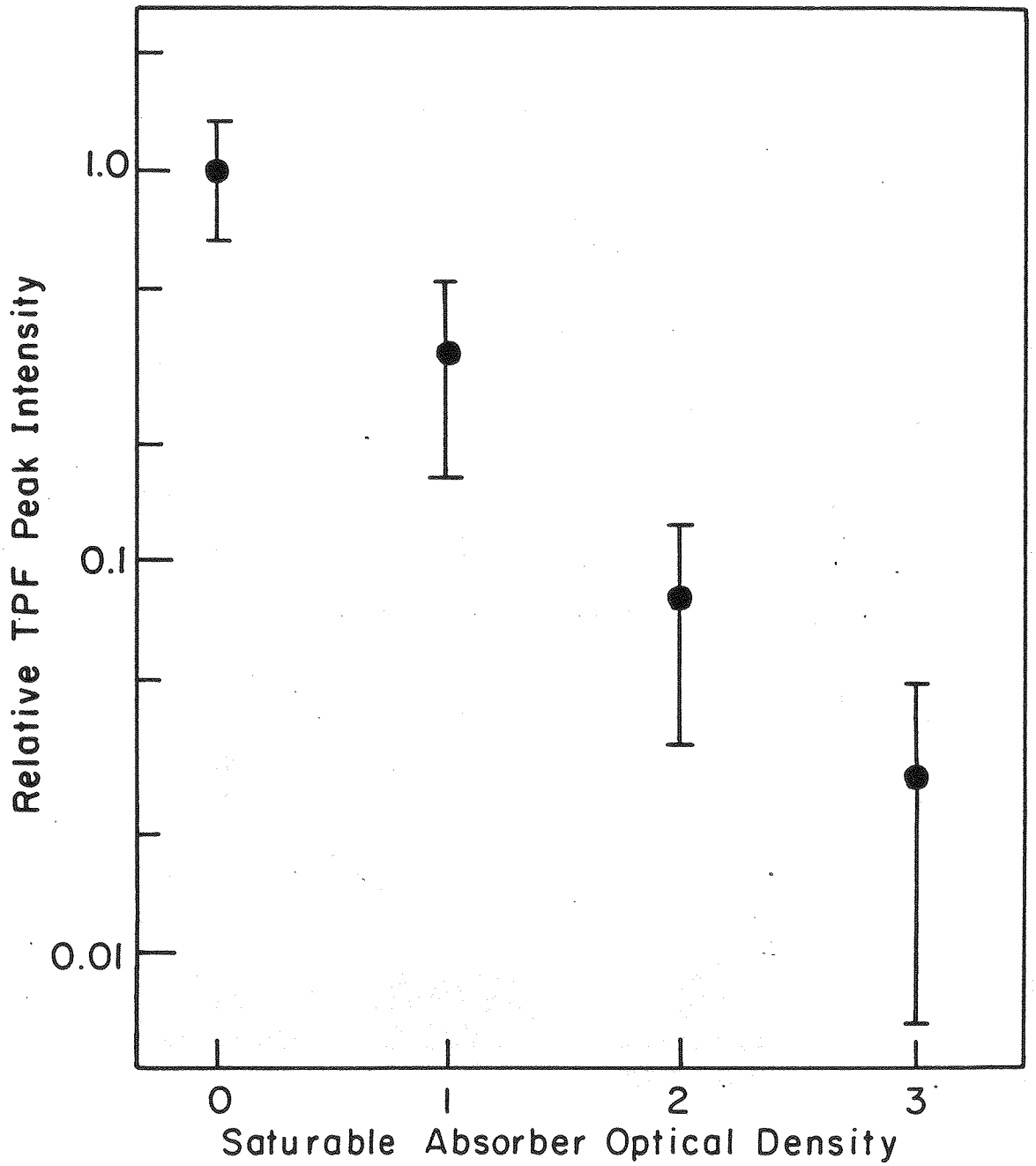
1mm
Edge of TPF cell
(a)
(b)
(c)
(d)
TPF Intensity Measured by Optical Multichannel Analyzer
Distance (mm) ⟶


Autocorrelation Width (mm)
2.2
2.0
1.8
1.6
1.4
1.2
Gaussian Pulse Width (psec)
12
10
8
0
1
2
3
Optical Density in 1 cm cell

Figur
3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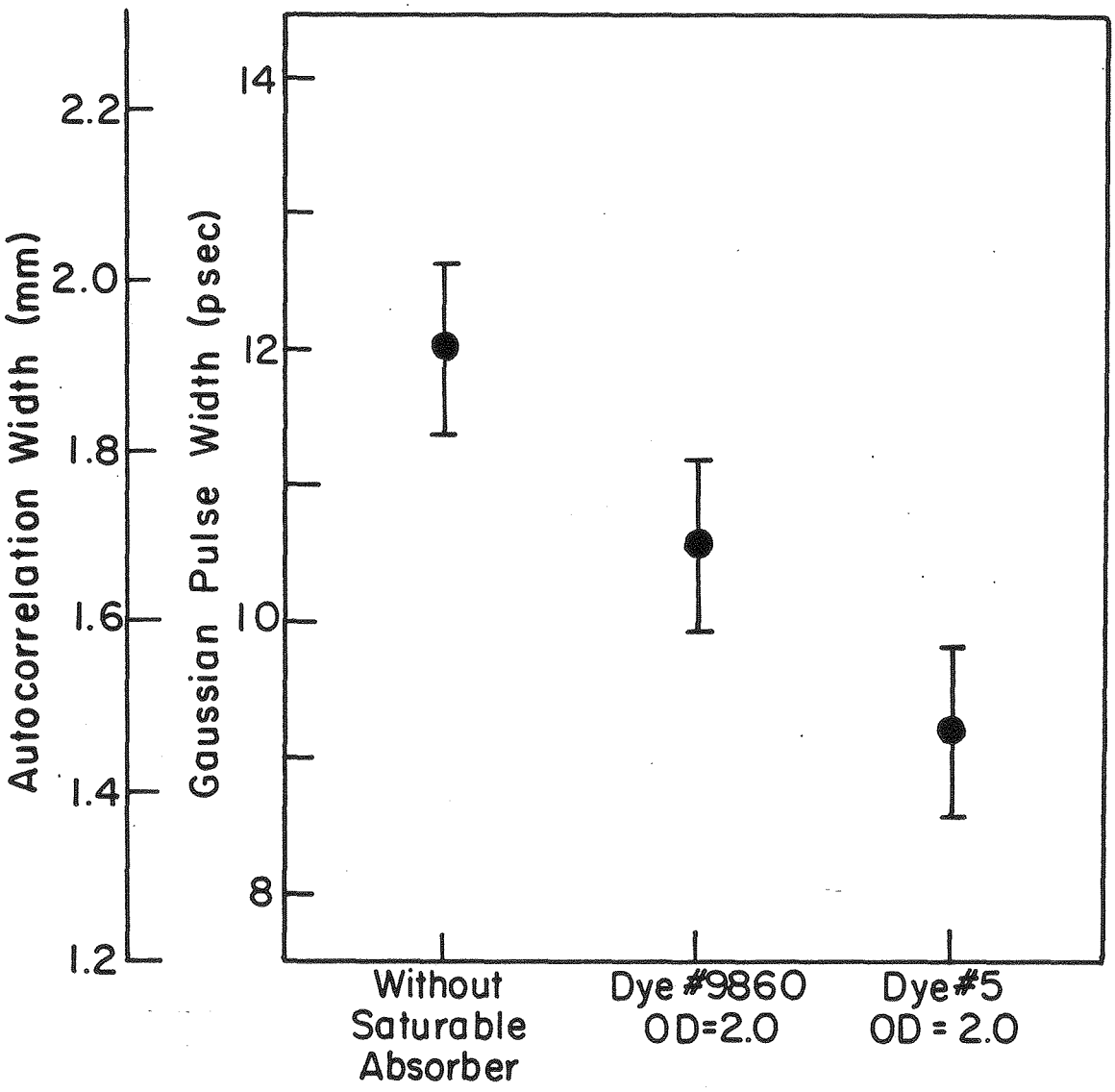
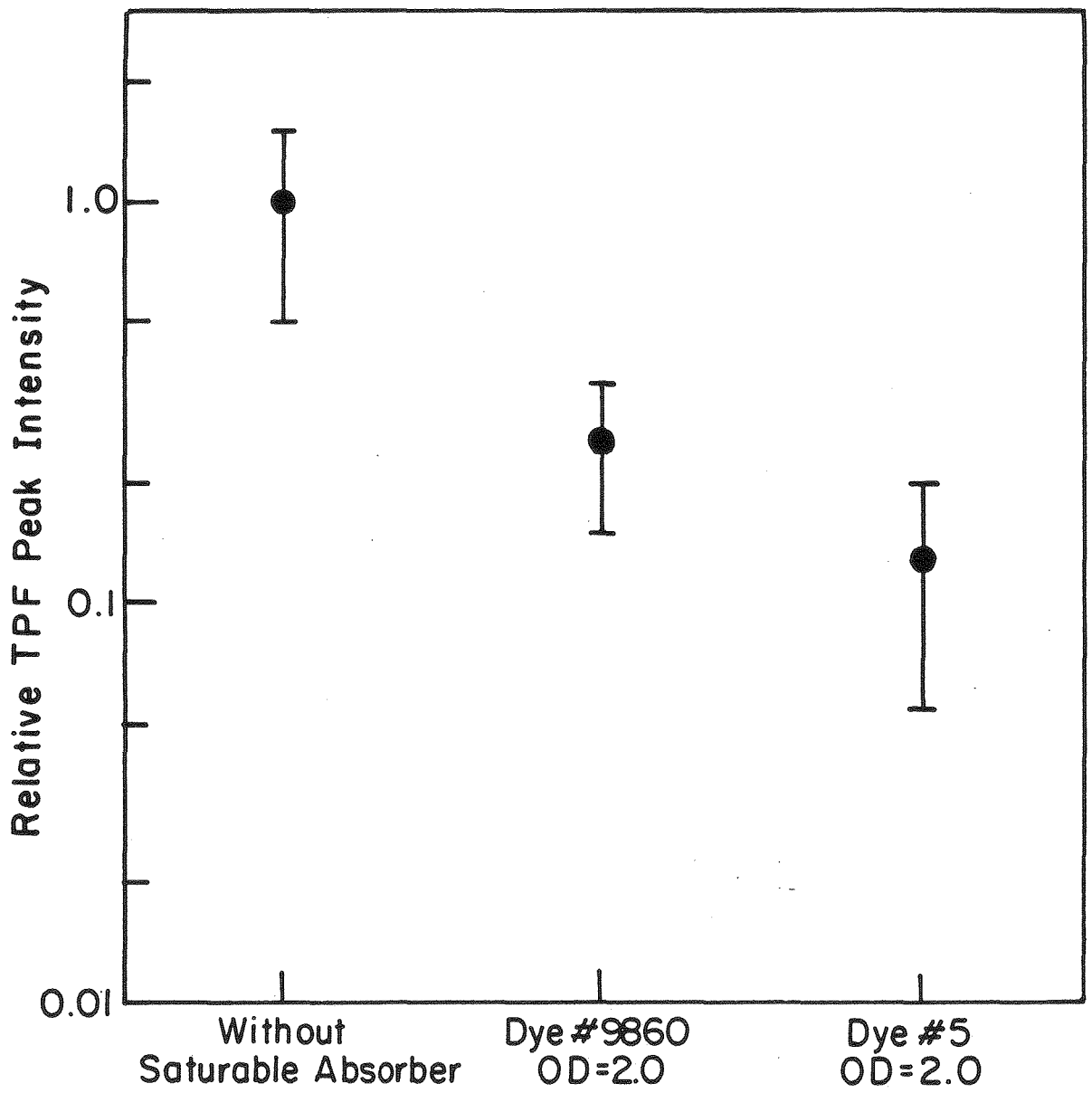
0.1
0.01
Relative TPF Peak Intensity
0
1
2
3
Saturable Absorber Optical Density


Autocorrelation Width (mm)
2.2
2.0
1.8
1.6
1.4
1.2
Gaussian Pulse Width (psec)
14
12
10
8
Without Saturable Absorber
Dye #9860 OD=2.0
Dye #5 OD = 2.0

Figur
5


Relative TPF Peak Intensity
1.0
0.1
0.01
Without
Saturable Absorber
Dye #9860
OD=2.0
Dye #5
OD=2.0