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1. AGENCY USE ONLY *(Leave blank)*	**2. REPORT DATE** May 15, 1992	**DATES COVERED** Final Report 3/15/89 to 3/31/92

4. TITLE AND SUBTITLE Subpicosecond Resolved Incipient Laser Damage	**5. FUNDING NUMBERS** ARO DAAL 03-89-K-0060
6. AUTHOR(S) W. E. Bron	
7. PERFORMING ORGANIZATION NAME(S) AND ADDRESS(ES) The Regents of the University of California, Irvine University of California Irvine, CA 92717	**8. PERFORMING ORGANIZATION REPORT NUMBER**
9. SPONSORING/MONITORING AGENCY NAME(S) AND ADDRESS(ES) U. S. Army Research Office P. O. Box 12211 Research Triangle Park, NC 27709-2211	**10. SPONSORING/MONITORING AGENCY REPORT NUMBER**

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SEP 11 1992
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11. SUPPLEMENTARY NOTES

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12a. DISTRIBUTION/AVAILABILITY STATEMENT Approved for public release; distribution unlimited.	**12b. DISTRIBUTION CODE**

13. ABSTRACT *(Maximum 200 words)*

Final report for an investigation of subpicosecond resolved incipient laser damage.

92 9 10 013

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14. SUBJECT TERMS	**15. NUMBER OF PAGES**
	16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	**18. SECURITY CLASSIFICATION**	**19. SECURITY CLASSIFICATION OF ABSTRACT**	**20. LIMITATION OF ABSTRACT**
UNCLASSIFIED	UNCLASSIFIED	UNCLASSIFIED	UL

NSN 7540-01-280-5500

Standard Form 298 (Rev 2-89)
Prescribed by ANSI Std Z39-18
298-102

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SUBPICOSECOND RESOLVED INCIPIENT LASER DAMAGE

FINAL REPORT

W. E. BRON

MAY 15, 1992

U. S. ARMY RESEARCH OFFICE

CONTRACT DAAL03-89-K-0060

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The overall goal of this line of research is to determine in detail laser induced transient dynamics in condensed matter in the presence of phonons and carriers at concentrations which exceed thermal equilibrium and approach the threshold for incipient damage.

Two major problems had to be solved before meaningful data could be obtained and a theoretical basis was achieved with which the observed transient dynamics could be understood.

The experimental problem of sufficient signal-to-noise to observe very fast optical transients in the presence of intense laser beams was solved through two unique laser systems. These laser systems consist of dual synchronously pumped and synchronously amplified dye lasers combined with the generation and kiloherz rate amplification of synchronized femtosecond and picosecond laser systems. Also important was the development of fast (1 KHz) analogue to digital converter needed to abstract the data and to regulate various components of the laser system. Specifically, this involved the construction of the 1 KHz computerized data acquisition system consisting of before and after sample photodiode beam detection and Raman signal detection, amplification, gated integration and a/d conversion. This pulse-by-pulse normalized scheme has been used to investigate the absorption properties of the GaP crystal necessary for this experiment.

The next step in the experiment is to investigate the evolution of localized damage by finite pulse trains ranging from single pulse effects to multi-pulse cumulative effects. This was realized by the incorporation of a beam shutter which can introduce a desired number of pulses into the crystal. At this point, investigation into the respective roles of phonons and excited carriers in the generation of localized damage was investigated.

An analysis of the early data on time resolved incipient laser damage indicated quite clearly that a detailed theoretical basis was required to understand the generation of excited carriers, through two-photon absorption (TPA), and cumulative damage produced and the subsequent cooling of the carriers. To facilitate a better understanding of the carrier cooling process, we incorporated results from a concurrent experiment conducted under different auspices. One aspect, requiring further study, centered on the cooling dynamics of optically induced hot carriers in GaP. A more complete understanding of the cooling process, from an initial excitation level of ~2eV above the indirect gap until recombination to the valence band, lead to a more complete understanding of the dynamics between hot carriers and optical phonons and their respective roles at the onset of damage.

At this stage of our understanding of the observed transients, it became possible to conduct a series of experiments designed to elucidate the following factors.

The experiments performed include (i) an investigation of threshold damage as a function of both incident laser pulse fluence and wavelength using high-energy pulse trains, (ii) damage evolution during the first 1000 incident pulses as a function of both incident laser pulse fluence and wavelength, and (iii) detailed studies of two photon absorption under the conditions of the two above mentioned parameters. A number of technical difficulties had to be overcome in order to produce a proper focal volume which would cause bulk damage in the crystal sample without producing damage to the surface and in the modulation of the pulse trains.

Experimental results of particular note were the observation of a rapid increase in the threshold for bulk damage as the duration of the incident pulse is decreased below ~500 femtoseconds and its total energy is kept constant. This increase in the damage threshold is in contradiction to observed effects of decreasing pulse durations in the picosecond and nanosecond time regimes. As the duration of the pulse is decreased from nanoseconds to picoseconds, the damage threshold is observed to decrease. We believe that the change at 500 fs may be due to a reduction of the free carrier absorption as the pulse duration decreases to the order of the carrier-carrier scattering time.

The observed increase in the damage threshold with decreasing laser pulse duration lead to a brief report submitted to the ARO. A direct copy of the report is included here as Appendix I. A more detailed report of the results currently in an early state of publication appears as Appendix II.

Laser Damage Refined

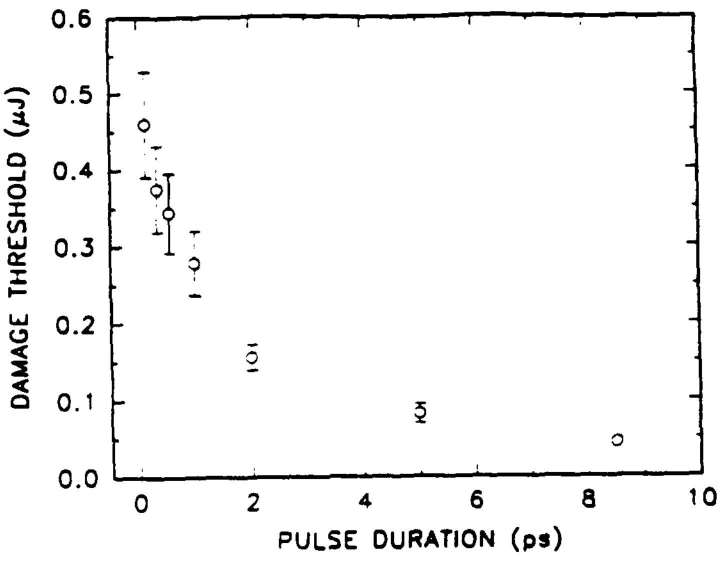
Researchers at the University of California at Irvine have found a not previously observed mechanism for laser induced bulk damage in compound semiconductors. The research group headed by Prof. W. E. Bron has applied, to measurements of the damage threshold, ultrashort duration laser pulses of duration of ~8 ps (10^{-12} s) down to ~135 fs (10^{-15} s). The laser pulses are produced in a unique, synchronous, multibeam, amplified laser system.

Much of the prior work in this field has subjected mostly large gap insulators to CW high intensity laser light. The strong, prolonged excitation leads to damage as a result of electric breakdown caused by impact ionization and electron avalanche processes.

It now appears that bulk laser damage can also be induced by ultrashort transient laser excitation of medium gap materials such as GaP. A proposed mechanism for this type of laser damage starts with carrier excitation through two-photon absorption followed by further excitation of carriers through one-photon absorption. The carriers eventually thermalize through phonon emission and, if the excitation is strong enough, it eventually leads to melting within the focal volume of the laser beam. It is predicted that the damage threshold, under these circumstances, increases as the laser pulse duration decreases below the onset for one-photon excitation, thereby decreasing the efficiency of the energy transfer from the laser pulse to the excited electrons, and lowers the possibility of sufficient energy transfer to cause local melting of the lattice. A second possible explanation involves strong self focusing of the laser beam as it propagates through the crystal. This mechanism, which is expected to be proportional to the inverse of the square of the laser pulse duration, does indeed agree with some of the observed data as indicated in Fig. 1. It is too early to definitely decide on one or the other, or both, as the effective mechanism. More research on this topic is required.

In any event, it is important to recognize that severe laser damage can occur not only in the presence of long duration (CW) high intensity laser fields, but can also occur at ultrashort, tightly focused laser pulses. Hardening of integrated circuitry subjected to ultrashort laser pulses will need to differ from that produced for electric breakdown.

This work was supported by funds from the Army Research Office under the program supervision of Dr. M. Ciftan. G. O. Smith and T. Juhasz are major coworkers on the project.



Damage threshold intensity as a function of laser pulse duration. The symbols are data points and indicate the statistical uncertainty of the measurements. The solid line is a best fit to the function a/t^2 with a ~ 0.06. The laser spot size is ~ 10 μm.

Bulk Damage in GaP Induced by Picosecond and Femtosecond Laser Pulses

G.O. Smith, T. Juhasz and W.E. Bron

Department of Physics, University of California, Irvine, CA 92717

Laser induced damage in optical materials has been widely investigated in the nanosecond time regime. It has been shown that bulk damage is due to localized melting and/or evaporation of the lattice when a sufficiently dense plasma is created either through collisional or multiphoton ionization. Extensive investigations on the picosecond and femtosecond time scales are still missing. We present results from an investigation into the dependence of the bulk damage threshold in GaP on pulse duration and cumulative effects due to multiple incident pulses. The incipient damage threshold is shown to increase rapidly for pulse durations of less than 700 fs.

The literature surrounding laser induced damage in optical materials is vast and covers a long series of possible mechanisms and a variety of experimental and theoretical methods with which to study the phenomenon. Some of these mechanisms include such effects as collisional (electron avalanche) and multiphoton ionization, hypersound generation during stimulated Mandel'shtam Brillouin scattering (SMBS), light pressure, electrostriction, and various types of thermal effects induced by absorbing inclusions and defects (thermoelastic stresses, thermal ionization, photoionization by radiation emitted by heated inclusions, etc.) [1,2]. Of the above mentioned mechanisms, collisional and multiphoton ionization, have been shown to be the most often present [3,4].

In dielectrics, with large energy gaps, multiphoton ionization is relatively weak leaving electron avalanche as the dominant mechanism for damage [4,7]. In the case of high photon energies and/or smaller gap materials (*e.g.*, semiconductors), and providing that multiphoton ionization is strong enough, the rate of excitation of electrons into the conduction band will exceed the corresponding rate for collisional ionization, and thus will dominate as the mechanism for laser induced damage [2]. Moreover, as the laser pulse duration decreases, the time available for avalanche ionization decreases. This limitation does not apply for multiphoton ionization which is dependent only on the laser field intensity.

The investigation to be discussed here is carried out using a synchronously amplified dye laser system operating at a repetition rate of 1 KHz [8]. Studies of the dependence of the damage threshold on pulse duration use pulses from a synchronously pumped, group velocity dispersion compensated, passively modelocked femtosecond dye laser which is synchronously amplified in a two-stage dye amplifier chain pumped by a Nd:YAG regenerative amplifier [9]. Pulse trains of 100 to 1000 pulses are focussed, via a 10x microscope objective, into the bulk of a 7 mm long, high purity ($<10^{17}$ cm^{-3}) [10] sample of single-crystalline GaP. Pulses of 135 fs to 8 ps in duration and 10 nJ to 4 μJ in energy are used. The pulses are focussed to a spot size of ~5 μm. The short working distance and tight focussing of the microscope objective causes the pulse irradiance to be sharply peaked over a distance of ~200 μm on either side of the beam waist. By measuring the pulse energy both before and after transmission through the sample, the dependence of the damage threshold and accumulated damage rate on pulse duration and pulse irradiance is determined. The sample is considered damaged if, over 1000 incident pulses, the percentage of light scattered from the damage volume exceeds 10% of that from low level irradiance. Additional information on the damage is obtained from diffraction patterns which arise from scattering of the beam at the edges of the damage volume. In addition, a visual inspection is made in order to verify that damage has, in fact, occurred. The same criterion is used to determine the number of incident pulses required for damage.

The experiment consists of two sets of measurements. In the first of these measurements, the bulk damage threshold is determined for pulse durations ranging from 135 fs to 8 ps. In the second set of measurements, the number of pulses necessary to achieve damage, during a train of 1000 pulses, is determined for energies ranging from 10 nJ to 4 μJ per pulse. The number of pulses necessary to achieve damage is empirically determined as the point at which the average transmitted energy decreases below 90%.

Figure 1 displays typical values for the normalized transmitted energy per pulse as a function of the number of pulses incident on the sample. Figure 1(a) indicates a train of 1000 pulses whose intensities are insufficient to cause damage. The transmitted pulse energy is divided by the incident energy for each pulse in order to reduce the noise associated with pulse to pulse fluctuations. Figure 1(b) indicates a pulse train with sufficient energy to cause damage in the crystal. The amount of transmitted light through the sample decreases rapidly during the initial stages of damage, followed by a saturation after ~500 pulses.

Figure 2 displays the irradiance needed to cause bulk damage as a function of the duration of the incident pulse. The threshold slowly increases until the pulse duration decreases to ~700 fs, below which the threshold increases rapidly. Figure 3 represents results on the number of incident pulses necessary to achieve damage verses the pulse irradiance for both 580 nm and 590 nm light. Both sets of data indicate an increase in the number of pulses necessary for damage as the pulse irradiance decreases.

It is generally accepted that the formation of a damage site arises when a sufficiently dense plasma is created (~10^{18} cm^{-3}) such that Joule heating of the strongly colliding electron gas, which is driven by the laser field, becomes so large that internally localized melting and/or evaporation takes place [4]. In our experiment, self-focussing of the beam is observed for pulse irradiances above the threshold for damage. Visual inspection of the damaged areas of the crystal indicate that the location of the damage site lies at the beam waist, when the pulse intensities at, or slightly larger than, the threshold for damage. As the pulse irradiance is increased, periodic damage filaments are observed along the beam axis with starting positions which move away from the beam waist towards the incident face of the crystal.

The two primary mechanisms for self-focussing, encountered for pulses on the nanosecond and subnanosecond time scale, are that of electrostriction and the nonlinear response of bound electrons. Electrostriction occurs under laser irradiation as the net electrostrictive force at any point is proportional to the square of the electric field. Thus, a radially symmetric beam will lead to a radially symmetric stress with an associated change in the refractive index leading to self-focussing. The acousto-optical interaction, therefore, involves a radially propagating compression wave driven by the intensity gradient of the laser field [2]. Since the time necessary for the acoustic response to develop is of the order of 10^{-10} s, we eliminate this as a possible mechanism for pulses of picosecond and shorter durations. Self-focussing due to nonlinear electronic response arises through changes in the nonlinear index of refraction with intensity which, in turn, is proportional to $\chi^{(3)}$. The response time of this mechanism is sufficiently short to be present for pulse durations in the femtosecond regime. Since the pulse durations used in our experiment are much shorter than the time needed to establish electrostriction, self-focussing due to nonlinear electronic response should be the dominant mechanism in our experiment.

Self-focussing has associated with it a periodic filamental damage structure [2]. We do not observe any periodic damage at laser irradiances up to, and at, the damage threshold. What damage we do observe occurs at the vicinity of the beam waist. It remains, therefore, to be determined whether or not a self-focussing mechanism exists only at the beam waist which causes the local energy density to reach threshold values, or whether or not, the incident laser field, without further focussing, is at the threshold value.

If the mechanism for damage at threshold irradiances is considered to be self-focussing, it would be expected that the threshold for damage would decrease as the pulse duration decreases. The decrease in the threshold would be due to the increase in the peak irradiance and thus a decrease in the self-focussing radius. This is clearly not the case as indicated in Fig. 2 and, therefore, we conclude that self-focussing is not the dominant mechanism responsible for the pulse duration dependence of the damage threshold. Furthermore, we believe that the change in the damage threshold with pulse duration is due to the dependence of free carrier absorption of the incident laser field on the duration of the incident pulse.

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Fig. 1. The normalized transmitted energy per pulse verses the number of pulses incident on the sample in the case where: (a) the pulse irradiances are insufficient to cause damage; and (b) the pulse irradiances are sufficient to cause damage.

Fig. 2. The pulse irradiance needed to cause damage verses the duration of the incident pulse.

Fig. 3. The number of incident pulses necessary to achieve damage verses pulse irradiance for 580 nm (closed triangles) and 590 nm (open circles).

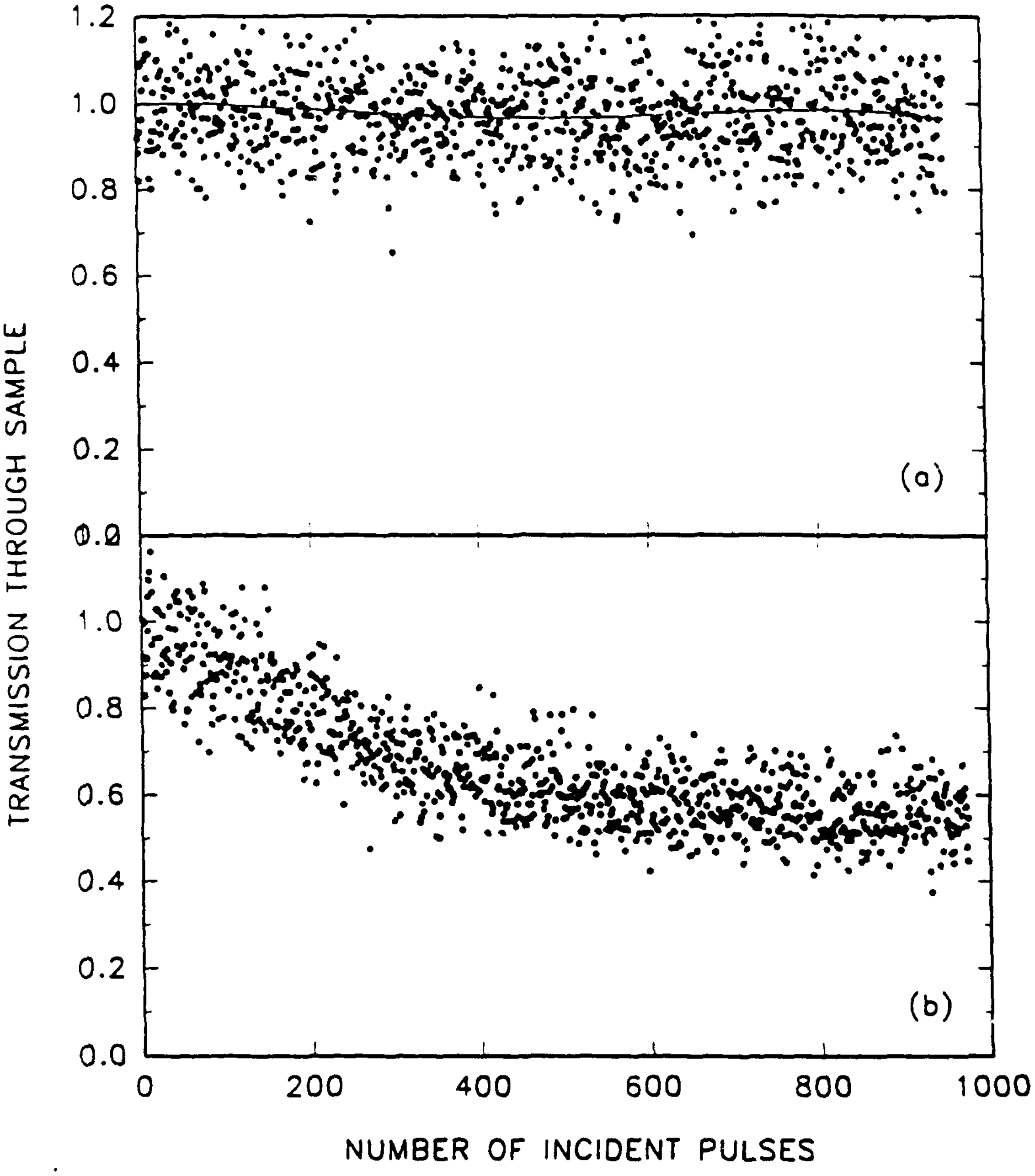
TRANSMISSION THROUGH SAMPLE
NUMBER OF INCIDENT PULSES
(a)
(b)
0.0
0.2
0.4
0.6
0.8
1.0
1.2
0
200
400
600
800
1000

FIG. 1

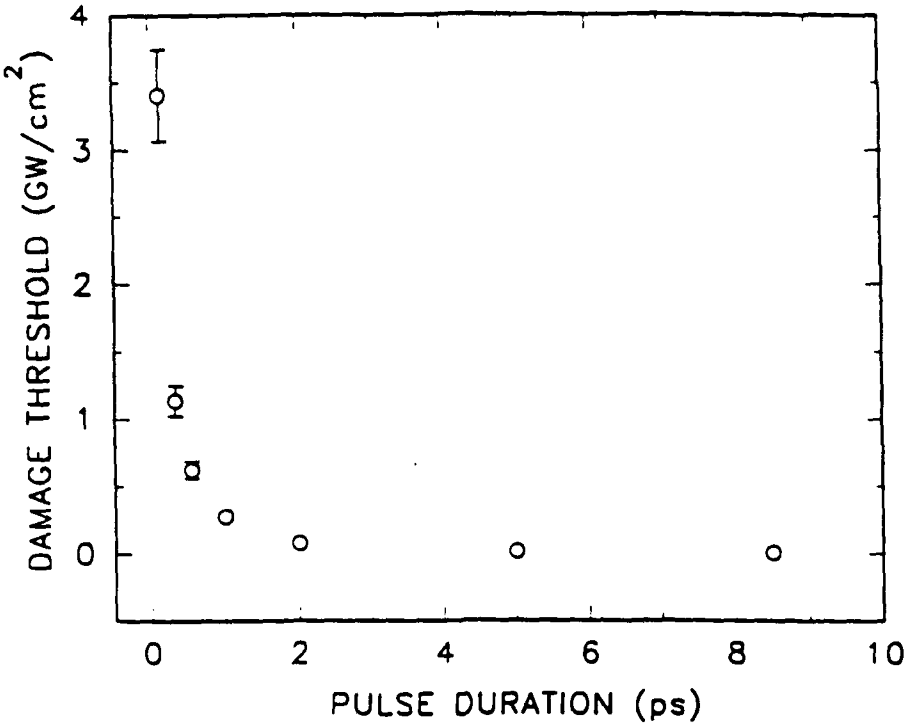

FIG. 2

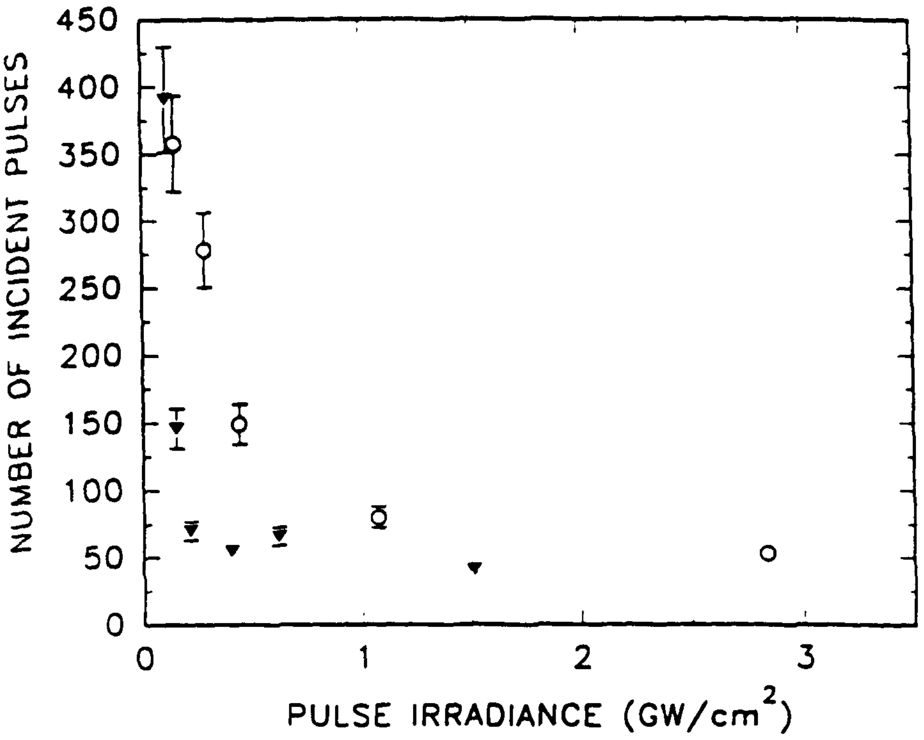
450
400
350
300
250
200
150
100
50
0
NUMBER OF INCIDENT PULSES
0
1
2
3
PULSE IRRADIANCE (GW/cm^2)

FIG. 3

Published Papers

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