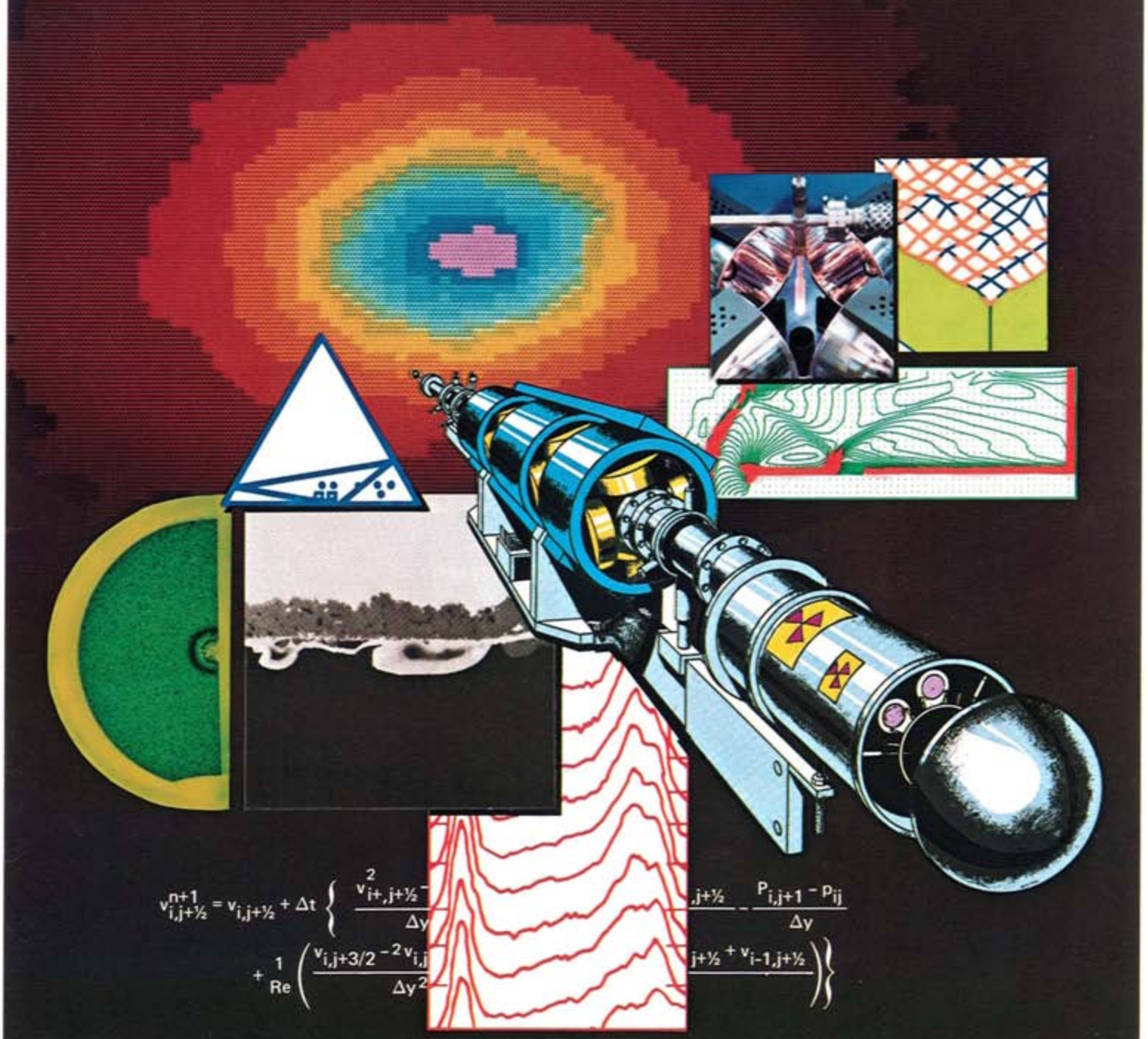
Energy and Technology Review

Lawrence Livermore Laboratory
November 1979

BASIC ENERGY SCIENCES I



$$v^{n+1}_{i,j+½} = v_{i,j+½} + \Delta t \left\{ \frac{v^2_{i+,j+½} - \quad ,j+½ - \frac{P_{i,j+1} - p_{ij}}{\Delta y}}{\Delta y} + \frac{1}{Re}\left(\frac{v_{i,j+3/2} - 2v_{i,j} \quad j+½ + v_{i-1,j+½}}{\Delta y^2} \right) \right\}$$

Absorptivity and damage thresholds in laser glass

The operating flux levels of lasers used for inertial fusion research are set by the damage thresholds of bare polished glass surfaces and optical thin films. Improved surfaces and films, with increased damage thresholds, would allow immediate increases in both laser power and laser energy. We have been conducting a vigorous effort to increase the damage thresholds of surfaces and films. To do so, we must understand the surface absorption and the relationship between absorptivity and damage threshold. As a first step, we have developed a sensitive photoacoustic technique for measuring surface absorption and have made absorption measurements using both short-pulse and constant-wave lasers. We have also measured absorption during short-pulse laser-damage experiments and have found an apparent increase in absorptivity at the damage threshold.

The study of optically induced damage to laser components comes naturally to the laser fusion program: much of our progress in achieving the requisite beam intensities for fusion research is due to our development of ways to prevent laser damage, and further improvements in performance would automatically accompany still more effective damage-prevention methods. Hence we are actively pursuing a theoretical and practical understanding of the mechanisms by which intense light can damage glasses and thin films.

Our unique facilities for studying optical damage include two large neodymium-glass lasers operating at 1.06 μm, one with pulse durations of 100 to 1000 ps and a maximum energy level of about 1 J and the other with pulse durations from 1 to 100 ns and an output power of about 1 GW, or 1 J at 1 ns and 100 J at 100 ns. Harmonic generation also provides pulses at 532, 355, and 266 nm. Our instrumentation enables us to measure pulse energy to within ±3%, absolute integrated energy density in a given plane to within ±5 to 7%, and absolute intensities to within ±10 to 15%.[1]

We originally performed the beam profiling essential to energy flux or intensity measurements by an accurate but cumbersome photographic technique. Instead, we now use a computer-controlled Vidicon profiler,[2] which provides real-time beam analyses and greatly speeds data acquisition. Figure 1 is a visual display generated by this equipment.

To detect and measure the extent of laser-induced damage, we examine exposed surfaces by Nomarski microscopy.[1] We have simplified this technique by constructing a device to move the irradiated surface into the focal plane of the microscope. This modification enables us to examine an area before irradiation, photograph it if necessary, move it into the beam and irradiate it, and then return it to the microscope for postirradiation evaluation.

Surface roughness and damage thresholds. In our first experiments, we looked for a correlation between the surface roughness of laser glass and its damage threshold.[3] We obtained silica samples of four different surface roughnesses by varying the duration of the final lapping with cerium oxide abrasives. We determined damage thresholds for these samples using 150-ps pulses, and the Air Force Weapons Laboratory independently tested them with 40-ns pulses. They observed a striking dependence on roughness, but

our data showed that roughness was relatively unimportant in setting 150-ps damage thresholds. We have since tested a variety of silica sufaces of varied roughness and determined that, for pulse lengths in the 0.1- to 3-ns range, typical of those used in laser fusion experiments, damage thresholds cannot be improved by conventional lapping once normal optical surfaces (root-mean-square roughness less than 4 nm) have been achieved.

Absorptivity and damage thresholds. We are now investigating the relationship between damage thresholds and surface absorption. The absorptivity of optical thin films, and of polished surfaces, is much higher than that of the bulk materials; we are attempting (1) to identify the source of this absorption and (2) to determine whether damage thresholds correlate with surface absorption levels.

We usually measure the absorptivity of thin films by laser calorimetry. Our present data indicate that if the absorptivity of a film 50 to 1000 nm thick is as high as 1000 millionths of the incident flux, the damage threshold will be low. When the absorptivity falls below 100 millionths, we no longer observe a correlation between absorption and damage threshold.

This correlation may fail for reasons not yet known to us, or it may simply appear to fail because laser calorimetry is unable to measure absorption accurately at such low levels. To investigate the latter possibility, we needed an independent method for measuring absorptivity. Therefore, we have developed a photoacoustic technique.[4]

When a sample absorbs energy from a laser pulse, some of this energy appears as a sudden heat pulse that stresses the material and generates a shock wave. This shock wave travels at the speed of sound in the material, usually without appreciable attenuation except at some material boundary, and can be picked up with a piezoelectric transducer.

To observe these shock waves, we bond a small, disk-shaped piezoelectric transducer to the rear surface of the sample to be irradiated and connect the transducer to an oscilloscope. Photographing the oscilloscope trace provides a permanent record from which we can directly read the magnitude of the photoacoustic signal.

This method can be used with both short-pulse and constant-wave lasers. In both cases there may be an additional signal from scattered light striking the transducer. For short-pulse lasers, this second signal causes relatively little confusion, partly because it is usually very weak compared with the primary

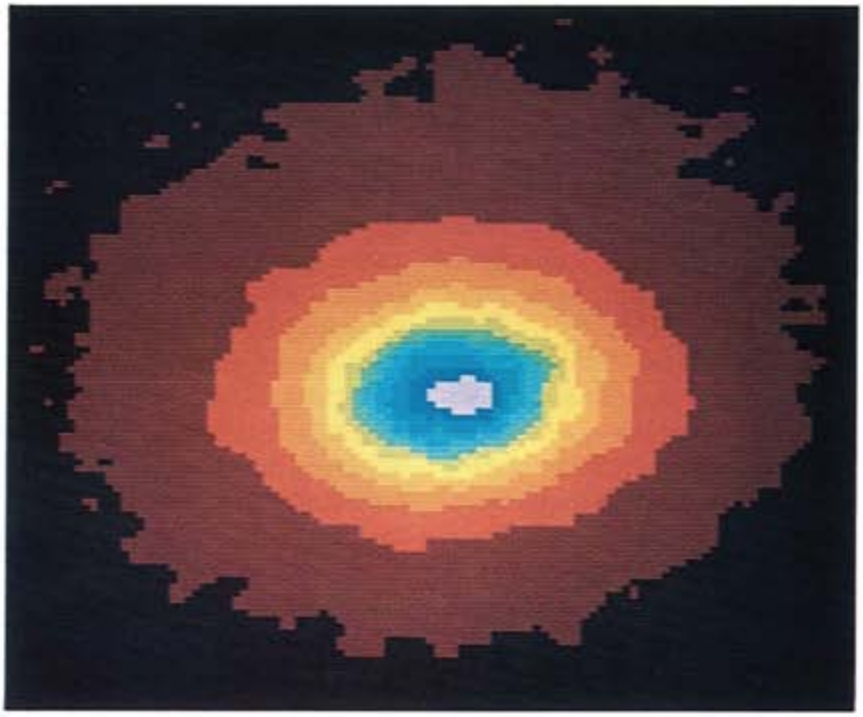


Fig. 1 *A laser-beam profile displayed by our computer-controlled Vidicon system. Each color contour represents a 6% change in energy flux. This system provides real-time beam analyses, computing the energy flux to within 10% immediately after each firing of the laser, and greatly expedites data acquisition.*

shock wave and partly because it always arrives early. The scattered-light signal usually dies away completely before the shock wave arrives.

For constant-wave lasers, however, the scattered-light signal arrives at the same time as the acoustic signal and must be subtracted from the total signal. To do so, we use an additional transducer, suspended near the sample. Since the second transducer has no contact with the sample, its signal comes entirely from scattered light and may be subtracted from the first transducer's signal to yield the true photoacoustic signal. To improve the signal-to-noise ratio, we use an electromechanical chopper to modulate the laser beam, and we analyze the resulting signal with phase-sensitive detectors tuned to the chopping frequency.

Absorption measurements. With our photoacoustic instrumentation, we have measured the absorption effects, below the damage threshold, of both long and short laser pulses. Such data validate our measurement technique and provide a base for understanding the nature of surface absorption in dielectric materials.

For constant-wave lasers, we have measured absorptions in the 100-millionths range with an accuracy of ±10%. For absorptions about one-fifth as large, the accuracy of our measurements was ±25% for high-reflectance films and ±50% for antireflectance films. This piezoelectric measurement, although its sensitivity is somewhat impaired by the need to subtract the scattered-light signal, can be applied more quickly and easily than laser calorimetry and is quite effective.

For short-pulse lasers, we measured absorption coefficients with pulses in the 1- to 50-mJ/cm^2 range, far below the damage threshold. Table 1 compares short-pulse absorption values measured by photoacoustic techniques with those measured by laser calorimetry. For test specimens, we took two samples of dielectric high-reflectance films—one for each measurement technique—from each of three coating runs and two identically polished pieces of bare silica substrate. Since our piezoelectric system was uncalibrated, we took the 120-millionths sample (sample 2) as a reference point. Although the absorption ranges over three orders of magnitude in these measurements, the agreement between the photoacoustic and the calorimetry results is

Table 1 *Optical absorptivity measurements, showing the degree of agreement between photoacoustic and calorimetric measurements. Specimens tested by the two techniques were similar but not identical, and this difference alone may be enough to account for most of the disagreement between the two sets of measurements.*

	Absorption, millionths of flux at 1.06 μm	
Sample[a]	**Laser calorimetry**	**Photoacoustic measurements**
1	13 000 ± 10%	6 000 ± 2%
2[b]	120 ± 20%	120 ± 2%
3	14 ± 50%	25 ± 10%
4	40 ± 30%	28 ± 10%

[a]Samples 1 through 3 were SiO_2–TiO_2 high-reflectance films; sample 4 was an uncoated silica substrate.
[b]Photoacoustic measurements were normalized by the measurement on this standard specimen.

reasonable, especially since the measurements involve two different sets of samples.

Our results indicate that, for short laser pulses, we can easily measure absorptions of a few tens of millionths with the pulsed piezoelectric method, and that we can attain a measurement repeatability of ±5%. This technique is as sensitive as laser calorimetry, even in its present state of development.

Damage-threshold measurements. For the other portion of our work, studying the correlation between absorptivity and damage threshold, we have conducted photoacoustic damage studies on a number of dielectric high-reflectivity films on silica substrates. Figure 2 presents typical results for one of these samples, irradiated with 1-ns, 1.06-μm pulses. Acoustic signals are observable over the energy-flux range from 0.2 to 20 J/cm^2. The vertical dashed line at 5.5 ± 0.5 J/cm^2 indicates the damage threshold determined by Nomarski microscopy.

Below the damage threshold, the photoacoustic signal increases quite linearly with the radiant energy flux. At the damage threshold the photoacoustic signal abruptly doubles or triples. For energy fluxes above 6 J/cm^2, the photoacoustic signal increases exponentially according to the fourth (±1) power of the incident energy flux.

We believe this large increase in acoustic signal at the threshold flux is not simply the noise of microcracking and fracturing that normally accompany laser damage. Similar experiments with metallic films show no comparable increase in the photoacoustic signal even when there is considerable damage. Apparently we are observing a sudden increase in optical absorptivity in the dielectric films, an increase that metallic films do not share.

The sharp increase in photoacoustic signal at the damage threshold in dielectric films may also provide an additional benefit: an opportunity for on-line, automatic monitoring of optical components in a high-energy laser system. We can easily attach a photoacoustic transducer to each of the amplifier disks, lenses, and polarizers in the system and put them all under electronic surveillance. Any shot-connected photoacoustic signal above a predetermined level would trigger an alarm to warn of possible optical damage.[5]

Summary. Photoacoustic measurements provide several important advantages, both for the measurement of low absorptivity in optical materials and thin-film

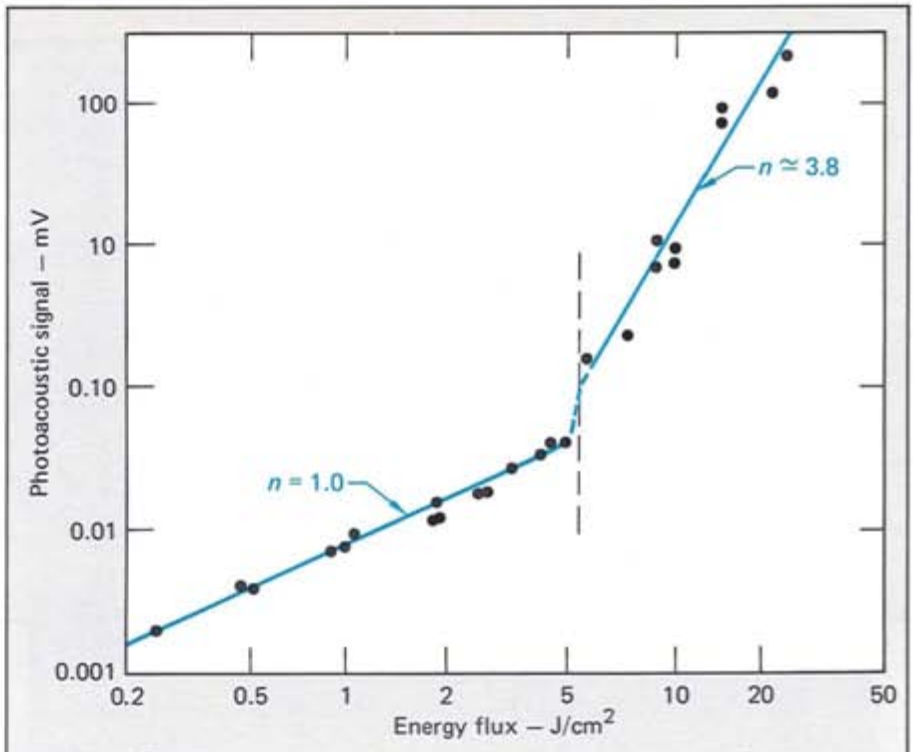


Fig. 2 *Variation of the photoacoustic signal as a function of the incident energy flux of laser light on a dielectric high-reflectance film. The dashed vertical line indicates the laser-induced damage threshold as determined by Nomarski microscopy. Absorption was linear (slope n = 1) for nondamaging flux levels but nonlinear (slope n ≃ 3.8) for fluxes above the threshold. This change suggests that absorptivity actually increases at the damage threshold.*

coatings and for the detection and study of laser-induced damage. The technique is as sensitive as laser calorimetry and much easier to apply. It can be used with both high-energy short-pulse lasers and less expensive constant-wave or low-energy short-pulse lasers. We plan to extend our photoacoustic measurements to other studies of laser materials, such as measurement of the radiative quantum efficiencies of neodymium glasses and the nonlinear absorption of ultraviolet in optical materials.

For further information contact Allan Rosencwaig (422-5374).

Key words: absorptivity, optical; damage thresholds; laser-induced damage; photoacoustic effect; thin films, absorptivity.

Notes and references

1. D. Milam, "Measurement and Identification of Laser-Damage Threshold in Thin Films," in *Optical Coatings–Applications and Utilization* (Society for Photo-Optical Instrumentation Engineers, Bellingham, Washington, 1978) vol. 140, p. 11.
2. W. Lee Smith, A. J. DeGroot, and M. J. Weber, "Silicon Vidicon System of Measuring Laser Intensity Profiles," *Appl. Optics* **17** (24), 15 (1978).
3. For discussions of the results of this study, see D. Milam, W. Lee Smith, M. J. Weber, A. H. Guenther, R. A. House, and J. R. Bettis, *Laser Damage in Optical Materials*, National Bureau of Standards, Gaithersburg, Maryland, Special Publication 509 (1977), or D. Milam, "1064-nm Laser Damage Thresholds of Polished Glass Surfaces as a Function of Pulse Duration and Surface Roughness," in *Proceedings of the Boulder Damage Conference*, 1978, National Bureau of Standards, Gaithersburg, Maryland, Special Publication 541 (1978).
4. A. Rosencwaig, "Photoacoustic Spectroscopy," in *Advances in Electronics and Electron Physics*, vol. 46, L. Marton, Ed., (Academic Press, New York, 1978), p. 207.
5. P. M. Johnson, C. A. Tatro, T. C. Kuklo, and A. E. Brown, "The Use of Acoustic Emission as a Detector of Damage Threshold," in *Laser Fusion Program Semiannual Report*, Lawrence Livermore Laboratory, Rept. UCRL-50021-72-2 (1972), p. 134.

Infrared spectra of deuterium-tritium

Because of their possible use as fuel in fusion reactors, we are measuring the physical and chemical properties of frozen mixtures of tritium and deuterium. One of our principal quantitative analytical tools is infrared spectroscopy. Our studies so far show that frozen D-T is a complicated mixture of D_2, DT, and T_2; molecular DT may be a better candidate for a fusion-reactor fuel.

Most of our present schemes for harnessing thermonuclear fusion reactions involve gaseous deuterium–tritium (D–T) fuels. A possible alternative is to use frozen D–T, which can be shaped into advantageous configurations for the nuclear burn. Accordingly, we have begun to measure the physical and chemical properties of solid D–T mixtures.

A mixture of hydrogen and deuterium is stable indefinitely unless it comes in contact with a catalyst. A D–T mixture, on the other hand, contains its own catalyst. Tritium is radioactive; its beta particle provides ionization that promotes the reaction

$$D_2 + T_2 \rightleftharpoons 2DT.$$

Thus at equilibrium a vessel filled with deuterium and tritium contains a mixture of three compounds: D_2, DT, and T_2.

Each of these compounds freezes at a slightly different temperature in the vicinity of 20 K. When the mixture begins to freeze, the first solid contains a slightly higher percentage of tritium than the gas phase. This process continues to deplete the tritium in the gas phase until all the tritium is used up and the last solid to form is almost pure deuterium. The result is a solid layered like a cake. To study this material, we need an instrument that can measure the components in these layers.