

New Method for Exposure Threshold Measurement of Laser Thermal Imaging Materials

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A new method is described for measuring the exposure threshold of laser thermal imaging materials exposed by pulses from a laser with a Gaussian radial spatial profile. This method, termed the maximum spot method, involves placing the material in the focused beam of a repetitively pulsed laser and exposing a series of spots of different sizes by moving the material through and beyond the beam focus. The exposure threshold can be deduced by knowing the radius of the largest exposed spot and the pulse energy. The method is demonstrated on a laser ablation transfer film (LasermaskTM) and a direct imaging film developed by Presstek, Inc., for computer-to-plate imaging applications. It is shown that the new method, which is convenient and quick, gives the same results with fewer sources of experimental errors as conventional threshold measurement methods. The convenience of the new method permits systematic studies of the dependence of exposure properties on material properties or laser imaging conditions. As an example, the imaging threshold of Lasermask is measured as a function of laser pulse duration from 10^{-12} to 10^{-4} s.

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Introduction

In this paper we describe a new method for accurately measuring the exposure thresholds of imaging materials used in laser thermal imaging processes, that are exposed by laser pulses having a Gaussian (TEM_{00}) radial spatial profile. In studies of the fundamental mechanisms of these materials, it is useful to determine the dependence of exposure thresholds on parameters such as material composition, spot size, pulse duration, exposure wavelength, etc. Such studies are ordinarily tedious and time-consuming, whereas our new method makes them convenient and straightforward.

Most studies of laser imaging exposure thresholds¹ involve subjecting the imaging material to a series of optical pulses of increasing fluence. One then determines the minimum fluence that produces a satisfactory image. This fluence J_{th} is deemed the exposure threshold. Several possible sources of error exist in this method. Difficulties may arise in identifying the exact threshold level unless the material has an extremely sharp exposure threshold. Once the imaging material is deemed exposed, the incident fluence that produced the exposure must be accurately determined. The fluence is the energy per unit area. Measuring the threshold pulse energy E_{th}^1 is usually easy, but measuring the laser beam radius r_0 is often problematic. The problem is compounded because J_{th} depends on the inverse square of r_0 . Tightly focused laser beams ($\sim 10 \mu\text{m}$) are used in high-resolution imaging applications. With tight focusing, the laser spot size r_0 is small and hard to measure and the value of r_0 depends critically on the distance from the focusing objective to the absorbing layer of the imaging material. It may be necessary to correct for the refractive index of the imaging material. For these

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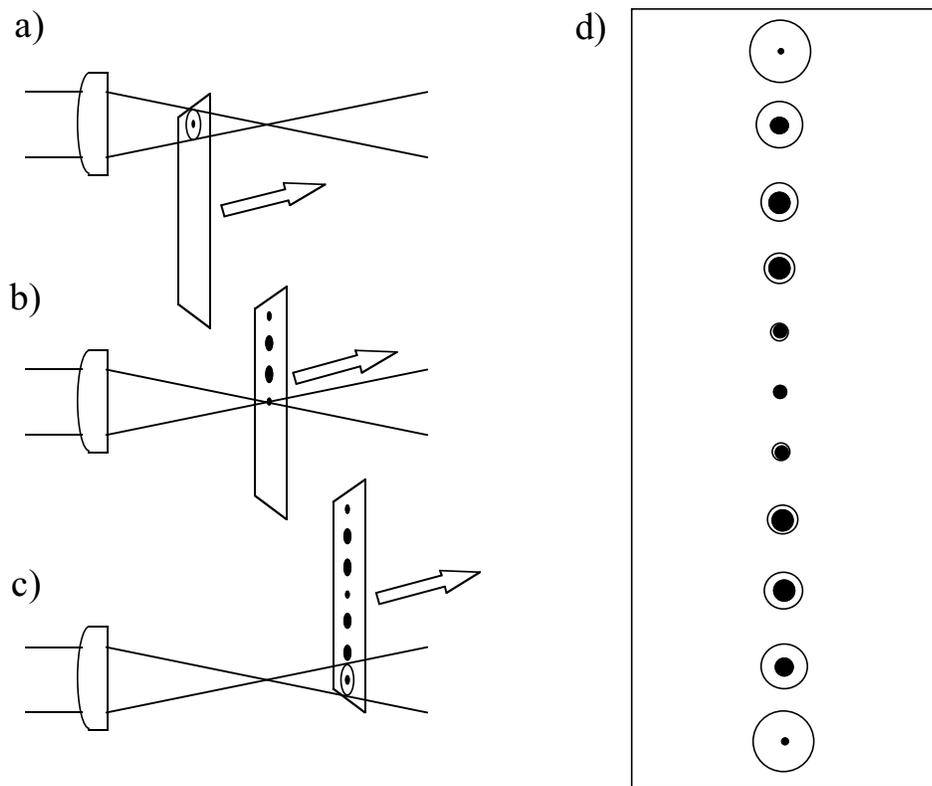


Figure 1. Schematic of the maximum spot method for measuring exposure thresholds. A strip of imaging material is placed in the beam of a repetitively pulsed laser with constant pulse energy E_p . In the figure, the open circles denote the $1/e^2$ contour of the Gaussian laser beam, with beam radius r_0 . The filled circles denote the exposed regions with radius r_s ; (a) The material is first placed where the beam is large so exposure does not occur. As the strip is moved through and beyond the focus (b) through (c) a series of spots that is exposed at different fluences. (d) From the record of exposed spots, the largest spot with spot radius r_s' is selected (the two largest spots are symmetrically arranged about a minimum) and the radius is measured. The largest spot is produced at the point of maximum efficiency. Knowing r_s' and E_p , Eq. 4(b) gives the exposure threshold J_{th} . From Eq. 4(a), the laser beam radius r_0' at this spot is exactly $\sqrt{2r_s'}$.

reasons, the determination of r_0 is usually the greatest source of error in threshold measurements.

When Gaussian profile laser pulses are used, exposure threshold measurements can be improved somewhat using a method originally described by Maydan.² The imaging material is exposed to a series of pulses with increasing intensity at and above imaging threshold. With a Gaussian pulse, the fluence decreases with radial distance from the beam center. When the fluence at the beam center reaches threshold, an infinitesimal exposed spot is created. For pulses above threshold, the radius of the exposed spot r_s increases with increasing pulse energy. A systematic measurement is made of pulse energy and spot size r_s . The data are fit to a theoretical model (Eq. 2) to extract J_{th} . In this method, J_{th} is determined over a range of pulse energies, greatly reducing the problem of identifying the threshold level. The method is time-consuming and does not solve the problem of errors in r_0 .

Our new method provides quick and direct measurement of the exposure threshold J_{th} without requiring an independent measurement of r_0 . In the rest of this report, we describe the new method, which we term the "maximum spot method." Experimental results are presented using two different laser imaging materials known to have sharp exposure thresholds. The results show the new method provides quantitative agreement with the Maydan method. The significance of the new method is demonstrated by making a measurement that is quite difficult with conventional tech-

niques, namely, the dependence of pulse fluence threshold over a wide range of pulse durations, from 10^{-12} to 10^{-4} s.

Theoretical

The idea of the maximum spot method is described in Fig. 1. A repetitively pulsed laser with a Gaussian laser beam profile is focused to a small spot with an imaging objective. A strip of the imaging material is held in the laser beam, away from the focal region [Fig. 1(a)]. The material is moved through the laser beam [Figs. 1(a) through 1(c)]. As the material is translated along the direction of beam propagation through the beam focus and beyond, it encounters successive laser pulses of the same energy E_p and duration t_p , but with different beam radii r_0 . When pulling the material through the beam, the material should be kept perpendicular to the beam, but the direction of travel should be at a slight angle from the direction of beam propagation [Fig. 1(c)], to ensure each laser pulse strikes a fresh spot of the material. With a little practice, one can expose a piece of imaging material in this manner in a few seconds. Figure 1(d) is a schematic of an exposed piece of imaging material. The open circles in Fig. 1(d) denote the $1/e^2$ intensity contour of the Gaussian beam located at radius r_0 from the beam center. The filled circles indicate the exposed spot in the imaging material with radius r_s .

The imaging material is first placed where r_0 is too large and the fluence too low to expose the film. As the film is

moved toward the focus, the fluence increases above threshold and a series of spots are exposed [Fig. 1(d)]. When r_0 is large [top of Fig. 1(d)], the laser energy is diffuse, only a tiny spot at the beam center is exposed and most of the laser energy is wasted. When r_0 is very small [center of Fig. 1(d)], the laser energy is concentrated at the beam center. The fluence at the beam center is far above threshold, and much of that laser energy is wasted. There is a point of maximum efficiency,^{2,3} where the available laser pulse energy E_p produces the largest possible exposed spot. The radius of the largest exposed spot is denoted r_s' . Knowledge of E_p and r_s' allows the exposure fluence threshold J_{th} to be determined without the need to measure the laser beam radius r_0 . In fact, the value of r_0 at the point of maximum efficiency, termed r_0' , is also determined by this method.

Consider a material with a sharp exposure threshold J_{th} independent of the size of the spot being exposed. This *spot size independence* is an important assumption critically examined below. For a laser beam with a Gaussian spatial profile, the fluence $J(r)$ delivered by a pulse with energy E_p and Gaussian beam radius r_0 ($1/e^2$ radius) is a radial function of the distance r from the beam center,⁴

$$J(r) = \frac{2E_p}{\pi r_0^2} \exp(-2r^2 / r_0^2) = J(0) \exp(-2r^2 / r_0^2). \quad (1)$$

When the fluence at the center of the beam $J(0)$ barely reaches threshold $J(0) = J_{th}$ and the film becomes exposed at a tiny region at the beam center. If the pulse energy is increased at constant r_0 , a larger spot is produced. The radius of the exposed spot r_s is given by the well-known relation,^{2-3,5}

$$\begin{aligned} \frac{r_s}{r_0} &= 0 & J(0) < J_{th} \\ \frac{r_s}{r_0} &= \sqrt{\frac{1}{2} \ln \left(\frac{E_p}{E_{th}} \right)} & J(0) \geq J_{th} \end{aligned} \quad (2)$$

In the Maydan method,² the exposed spot radius r_s is plotted as a function of the pulse energy E_p and the data are fit to a smooth curve defined by Eq. 2 to extract the threshold energy E_{th} . Then r_0 is measured and the threshold energy can be converted into a threshold fluence $J_{th} = 2E_{th} / \pi r_0^2$.

In the maximum spot method, the pulse energy E_p is held at a constant value and the beam radius r_0 is varied, producing a series of exposed spots with various spot radii r_s . The radius of the exposed spot r_s is always the distance from the beam center where the fluence $J(r)$ drops to the threshold value J_{th} . Thus,

$$J_{th} = \frac{2E_p}{\pi r_0^2} \exp\left(-\frac{2r_s^2}{r_0^2}\right). \quad (3)$$

Holding E_p and J_{th} constant, we vary r_0 to find the maximum value of r_s . That defines the point of maximum efficiency where r_s is a maximum r_s' . By differentiating Eq. 3 and then setting $dr_s/dr_0 = 0$, we obtain,

$$r_s' = \frac{r_0}{\sqrt{2}} \quad (4a)$$

and

$$J_{th} = \frac{E_p}{\pi(r_s')^2} \exp(-1), \quad (4b)$$

where r_0' is the Gaussian beam radius at the point of maximum efficiency.³ Equations 4(a) and 4(b) form the theoretical basis of the maximum spot method. If the pulse energy E_p and the radius r_s' of the largest exposed spot are known, Eq. 4(b) can be used to compute the threshold fluence J_{th} . In addition, Eq. 4(a) can be used to compute the Gaussian beam radius r_0' at the location where the largest spot was exposed.

In making threshold measurements with the maximum spot method, two cautions must be observed. First, if the pulse energy E_p is not large enough (or if the beam waist is not small enough), the imaging material is not exposed and obviously no result is obtained. However, if the laser pulse fluence is just the bare minimum that exposes the imaging material only at the beam waist, then the value inferred for J_{th} will be in error. If that should occur, the pulse energy should be increased a bit (or the beam should be focused more tightly). When the imaging material is then moved through the beam, two maxima in exposed spot size will be observed, one on either side of the beam waist [as in Fig. 1(d) and Fig. 4], because a point of maximum efficiency exists on either side of the waist. This double maximum is a sign that the pulse energy is in the right regime for accurate measurement of J_{th} .

Second, the exposure threshold of the imaging material might have spot size dependence. Because the value of J_{th} is determined using exposed spots of different sizes, such a dependence might be problematic for the method. A dependence of J_{th} on spot size would be caused by edge effects. For example, diffusion of heat away from the edge of the spot can cause spot size dependence. For a pulse of duration t_p , a characteristic thermal diffusion length λ_D exists in the imaging material. When the spot radius is small $r_s \leq \lambda_D$, thermal diffusion from the beam center to the edges can produce a spot size dependence of J_{th} . For sufficiently large spots $r_s \gg \lambda_D$, thermal diffusion along the radial direction would have essentially no effect on the exposed spot size and J_{th} would become independent of spot size.

A simple test can be made to determine whether J_{th} falls in the size-independent limit. The value of J_{th} should be determined at a few different values of pulse energy E_p . When E_p is smaller, the value of J_{th} derives from a smaller exposed spot. When E_p is larger, the value of J_{th} derives from a larger exposed spot. Thus, the assumption of spot size independence implies that the value of J_{th} measured by the maximum spot method should be independent of E_p , which can be empirically verified. For a given value of E_p , $\zeta = \sqrt{2E_p / \pi J_{th}}$ is the laser beam radius when the center of the beam just exceeds exposure threshold. In Fig. 2, we plot the theoretical relationship expected in a maximum spot experiment when J_{th} is independent of spot size. For a given ζ , when the beam radius r_0 is changed by moving the imaging material along the path of the focused beam, the exposed spot radius r_s will have a maximum value r_s' . For a material with a given J_{th} , when ζ is increased (by increasing E_p), a maximum spot radius is still observed, but the value of r_s' is larger.

Experimental

To demonstrate the maximum spot method, two quite different laser thermal imaging materials were studied,

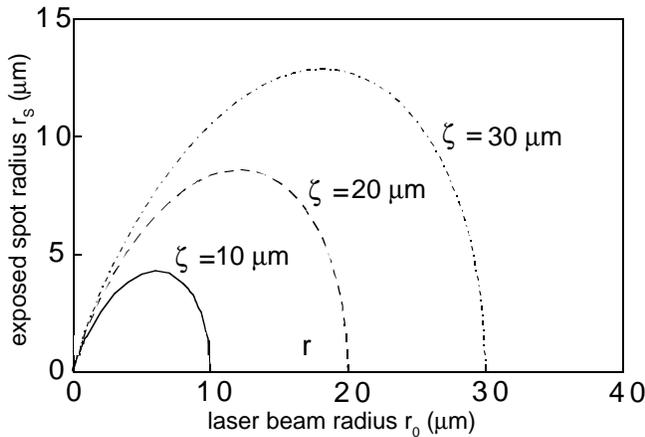


Figure 2. Theoretical plots of the dependence of the exposed spot radius r_s on the laser beam radius r_0 at constant pulse energy E_p for an imaging material with threshold fluence J_{th} that does not depend on spot radius. The parameter $\zeta = \sqrt{2E_p / \pi J_{th}}$ is the laser beam radius when the center of the beam just exceeds exposure threshold. For a particular value of J_{th} (that depends on the imaging material and possibly the pulse duration), the $\zeta = 10\text{-}\mu\text{m}$ curve would be obtained using a laser pulse of radius $10\text{-}\mu\text{m}$ and pulse energies E_p that cause the center of the beam just to exceed exposure threshold. Similarly, the $\zeta = 20\text{-}$ and $30\text{-}\mu\text{m}$ curves correspond to larger diameter beams with four and nine times the pulse energy used for the $\zeta = 10\text{-}\mu\text{m}$ curve. For a given value of ζ or E_p , there is a well-defined maximum r_s' of the exposed spot radius, which occurs at laser beam radius r_0' . For larger values of ζ , the

both of which were previously shown to evidence a sharp threshold for exposure by near-IR laser pulses. The first was a film (Lasermask) used in laser ablation transfer imaging. The Lasermask film has been described previously.^{5,6} It consists of a polyester substrate with an $\sim 1\text{-}\mu\text{m}$ -thick black coating, which is a suspension of graphite particle absorbers in an approximately 50:50 mixture of ethylcellulose and a phenolic resin. The Lasermask film was exposed from the substrate side. Ablation of the black coating left behind a clear spot in the film.

The second film is a model system for computer-to-plate imaging, similar to the commercial product used in the direct imaging PearlTM computer-to-plate system developed by Presstek, Inc. The direct imaging Presstek film was also described previously.^{3,7} It consists of a polyester substrate, an $\sim 30\text{-nm}$ -thick absorbing interlayer made of metallic titanium and titanium oxides, and a $2\text{-}\mu\text{m}$ -thick imaging layer of silicone polymer. The Presstek film was exposed from the substrate side as well.³ Following exposure, the film was cleaned with a soft cotton pad and some rubbing alcohol. After cleaning, a spot is produced where the silicone layer was removed.

The experimental apparatus was described previously.^{3,5} For most experiments, the exposure source was a continuously pumped, TEM₀₀ Nd:YAG laser (YAG denotes yttrium aluminum garnet) operating at $1.064\text{-}\mu\text{m}$ wavelength. Pulses in the 1 to 100 μs range were generated with an extra-cavity acousto-optic modulator, which sliced the pulses from the continuous laser output. When an intracavity acousto-optic Q-switch was used, giant pulses of 110-ns duration were obtained. Picosecond pulses (150-ps duration) used in a few experiments were generated using a similar laser equipped with an acousto-optic mode locker Q-switch and electro-optic cavity dumper.⁸ Subpicosecond pulses (0.5-ps duration) were generated by an amplified Ti:sapphire laser pulse at $0.769\text{ }\mu\text{m}$. Although the

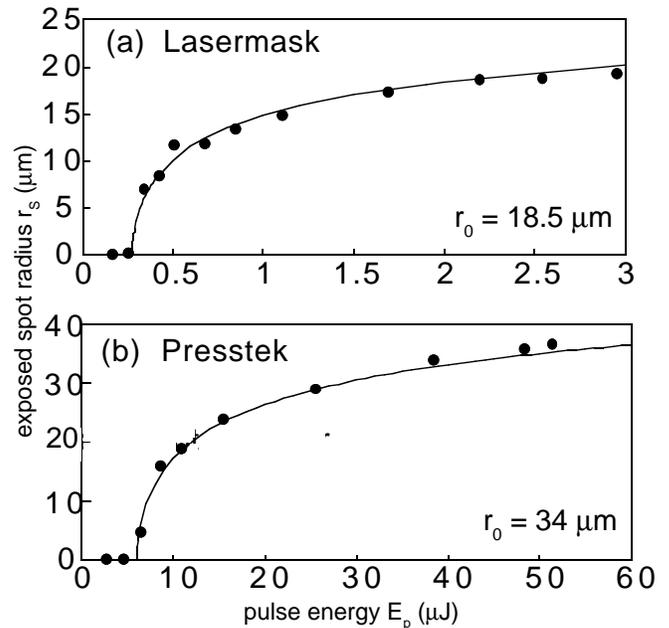


Figure 3. Measurements of exposure threshold using a conventional method, where the pulse energy is varied and the exposed spot size r_s is measured. The smooth curve is obtained by fitting the data to Eq. 2, by varying the exposure threshold energy E_{th} . To compute the exposure threshold J_{th} , an independent measurement of the laser beam radius r_0 is needed. The measured thresholds are given in Table I.

TABLE I. Threshold Measurements by a Conventional Method

Imaging material	Exposure pulse duration	Laser beam radius r_0	Exposure threshold fluence J_{th}
Lasermask ablation film	110 ns	18.5 μm	52 (± 5) mJ/cm ²
Presstek direct imaging film	10 μs	17.6 μm	340 (± 30) mJ/cm ²
Presstek direct imaging film	10 μs	34 μm	340 (± 30) mJ/cm ²

Ti:sapphire wavelength is different from the $1.064\text{-}\mu\text{m}$ wavelength of YAG used in other measurements, we do not believe that difference significant because the Lasermask film is black and its absorbance is almost exactly identical at both wavelengths. The radius of the exposed spot was determined using an optical microscope equipped with a video camera and computerized frame grabber and image analysis software. Typically, several strips of imaging materials were exposed and checked for the double maximum in exposed spot size. The largest spots on either side of the beam waist were identified, the radii determined, and an average threshold and a standard deviation calculated.

Results

Conventional Threshold Measurements. Using the Maydan method, the exposure thresholds of the two film materials were measured. The Lasermask ablation film was exposed using 110-ns-duration pulses, and the Presstek direct imaging film by 10- μs -duration pulses. These pulse durations are typical for commercial applications of these films.^{3,5} The Gaussian beam radius, which was determined using a knife-edge test,⁴ was $r_0 = 18.5\text{ }\mu\text{m}$ for the Lasermask measurements. To test for size-independence of J_{th} , the Presstek film was measured at two different beam radii, $r_0 = 17.6$ and $34\text{ }\mu\text{m}$. Figure 3 shows the results obtained with this method. The smooth curves

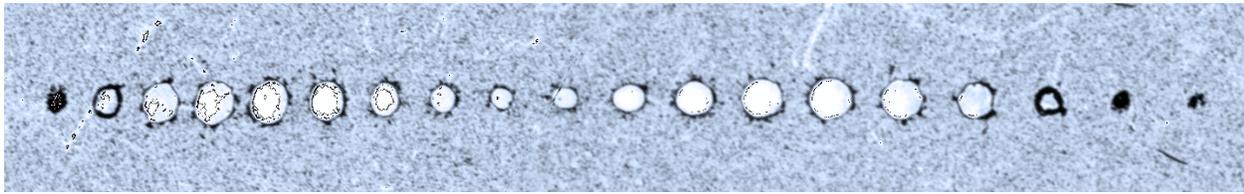


Figure 4. A strip of Lasermask film exposed to 110-ns-duration pulses from a Nd:YAG laser. To compute the exposure threshold, only the pulse energy (here $E_p = 7.7 \mu\text{J}$) and the radius r_s' of the largest spot are needed. The exposure thresholds determined by this maximum spot method are given in Table II.

TABLE II. Threshold Measurements by the Maximum Spot Method

Imaging material	Exposure pulse duration	Pulse energy E_p	Maximum spot radius r_s'	Laser beam radius r_0'	Exposure threshold fluence J_{th}
Lasermask ablation film	110 ns	7.7 μJ	29.8 μm	42.1 μm	51 (± 2) mJ/cm^2
Lasermask ablation film	110 ns	0.79 μJ	9.7 μm	13.7 μm	49 (± 2) mJ/cm^2
Presstek direct imaging film	10 μs	66 μJ	32.4 μm	45.8 μm	370 (± 30) mJ/cm^2
Presstek direct imaging film	10 μs	6.1 μJ	10.2 μm	14.4 μm	340 (± 30) mJ/cm^2

in the figure are fits to Eq. 2 obtained by varying E_{th} . The fact that Eq. 2 accurately fits the data over a range of spot sizes shows the exposure threshold is very sharp. The thresholds obtained by this fitting method are given in Table I. Most of the error in determining the threshold is attributed to error in determining the value of r_0 . The threshold for the Presstek direct imaging film is the same at both 17.6- and 34- μm spot sizes.³

Maximum Spot Threshold Measurements. An example of a maximum spot threshold experiment is shown in Fig. 4, which shows Lasermask film exposed by 110-ns pulses. At the left of the film strip, the laser beam radius is too large for the pulse to have any effect. Moving toward the right where the radius decreases and the fluence increases, at first small burned spots form, but the pulse fluence is too low to expose the film properly. Then exposed spots appear. A properly exposed spot has a light color, indicating removal of the black surface coating. The exposed spot radius increases to a maximum at the point of maximum efficiency. Then as the beam radius continues to decrease, the exposed spot size begins to decrease until it reaches a minimum at the center of the film strip. This minimum occurs at the beam waist. Continuing to the right, the spot radius again increases toward the second point of maximum efficiency. The symmetric double maximum in exposed spot radius indicates this measurement is being performed with fluences in the range needed to obtain accurate threshold values. The energy of the pulse E_p was 7.7 μJ . The radius of the maximum spot was $r_s = 29.8 \mu\text{m}$, which gives a laser beam radius $r_0 = 42.1 \mu\text{m}$ and an exposure threshold $J_{th} = 51 \text{ mJ}/\text{cm}^2$. Both types of films were studied at two different pulse energies, which differed by 1 order of magnitude. The results, summarized in Table II, show J_{th} is independent of spot size in the range studied.

In the maximum spot method, one possible source of error arises from determining the maximum spot radius from a finite number of exposures (Fig. 4), none of which is exactly the true maximum. However, Fig. 3 shows that the theoretical relationship between r_s and r_0 is relatively flat in the region of r_s' , so this problem is not serious. In practice, we found the largest source of error was actually measuring the maximum spot radius r_s' , which became most significant when r_s' was very small.

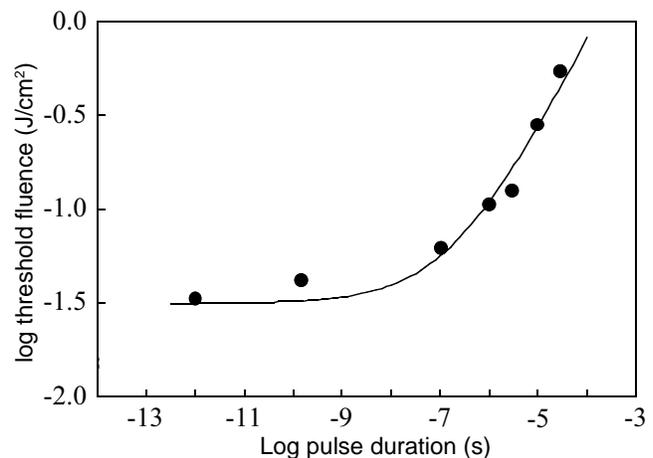


Figure 5. Pulse width dependence of exposure threshold for Lasermask film obtained using the maximum spot method with differently configured near-IR lasers, that produce various pulse durations. The threshold varies by a factor of ~ 20 for pulses between 0.5 ps and 30 μs duration.

Pulse Duration Studies. The dependence of exposure threshold on pulse duration t_p was studied for the Lasermask film over a range of pulsewidths spanning eight decades in time, $t_p = 0.5 \text{ ps}$ to 30 μs . For t_p greater than 30 μs , the film did not ablate properly. No existing laser can produce variable duration pulsewidths over such a broad range, so the measurements were made using three different lasers described above. The results are plotted in Fig. 5. It is seen the exposure threshold decreases by a factor of ~ 20 as the pulse duration is decreased from 30 μs to 100 ps. The smooth curve is a fit to a theoretical model of pulse duration dependence. The details will be discussed in a subsequent publication.⁹ It turns out that the less efficient imaging at longer pulse durations is due to the effects of thermal diffusion, which for longer pulses tends to reduce the peak temperature in the material.

Discussion

Comparison of Two Methods. Tables I and II show that exposure threshold measurements made by the two

methods agree within experimental error. Also the tables show the exposure thresholds are independent of spot size in the range studied. In our maximum spot measurements, we took the data with two different laser pulse energies which differed by a factor of 10. Consequently, the exposed spot size at the point of maximum efficiency varied by a factor of ~3, from approximately 15- to 45- μm radius. These conclusions should not be taken to indicate the threshold is independent of size below 15- μm radius, but they almost certainly do indicate the threshold is independent of size above 15- μm radius.

It generally takes us several hours for us to measure a threshold using the Maydan method. First we have to measure the laser beam radius r_0 . The knife-edge method we use is tedious, because many data points must be taken and fit to an error function, and it is difficult to be certain that the knife edge is precisely in the plane of the absorbing layer of the film. An accuracy of better than 1 μm is often required. Then the film must be exposed at many different intensity levels, and the pulse energy and exposed spot size must be measured at each exposure level. Finally Eq. 2 must be fit to the data to obtain J_{th} .

In contrast, in the maximum spot method the laser intensity is adjusted and a few trial exposures are taken to be certain the combination of focusing objective and laser pulse energy are in a reasonable range that gives the double maximum. A film is exposed and the largest spot is identified. Now only two measurements must be made, the energy of the laser pulse and the radius of the largest spot. No curve fitting is necessary. The pulse energy and spot size are merely inserted into Eq. 4(b) and the threshold determined. The entire procedure takes only a few minutes.

Significance of the New Method. The significance of the new method becomes clear when one contemplates systematic studies of exposure thresholds, such as the pulsewidth variation of the threshold shown in Fig. 5. In that type of measurement, the threshold must be measured with many different pulse durations, which in practice requires using several different lasers. Errors in determining the laser beam radius r_0 usually dominate threshold measurements by conventional techniques, and this measurement would require determining r_0 for each laser setup. In contrast, the maximum spot method takes a few minutes with each laser. Practically speaking, we

could never obtain reliable data of the type shown in Fig. 5 before we began using the maximum spot method.

Conclusions

A convenient new method has been demonstrated for measuring the exposure threshold of laser photothermal imaging materials exposed to optical pulses with a Gaussian radial intensity profile. Two quite different kinds of imaging materials were studied, which shows the method is quite general and would be especially useful in evaluating a series of various imaging materials with different material formulations. The method is predicated on having an exposure threshold independent of imaging spot size. However, our experiments show this is usually the case, and a simple test for spot size independence can be easily performed. The significance of the new method is demonstrated by obtaining a complete dependence of the exposure threshold on optical pulse duration, a difficult measurement that involves several different lasers to provide the needed range of pulse durations. \blacktriangle

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