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1993 Meas. Sci. Technol. 4 591

(http://iopscience.iop.org/0957-0233/4/5/008)

Meas. Sci. Technol. 4 (1993) 591–595. Printed in the UK

Determination of the laser-induced damage threshold of bulk polymer samples at 1.06 μm using the pulsed photothermal deflection technique

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Received 2 August 1991, in final form 7 January 1993, accepted for publication 15 January 1993

Abstract. A pulsed Nd–YAG laser beam is used to produce a transient refractive index gradient in air adjoining the plane surface of the sample material. This refractive index gradient is probed by a continuous He–Ne laser beam propagating parallel to the sample surface. The observed deflection signals produced by the probe beam exhibit drastic variations when the pump laser energy density crosses the damage threshold for the sample. The measurements are used to estimate the damage threshold for a few polymer samples. The present values are found to be in good agreement with those determined by other methods.

1. Introduction

The study of laser beam interaction has great practical value in addition to its importance from a fundamental standpoint. Laser-assisted etching of polymeric materials has proved to be a viable technique for photolithography (Dyer and Sidhu 1985, Srinivasan and Bodil Braren 1989) in the fabrication of microelectronic circuits. Some transparent polymers are applied as nonlinear optical materials (Lipscomb *et al* 1981, Milam 1977). Also, conventional laser materials such as glasses and crystals are increasingly being replaced by transparent polymeric materials as optical components in high-power laser systems (Dyumaev *et al* 1983).

Under normal conditions, materials suitable for applications in high-power lasers should have a high damage threshold. This necessitates evaluation of the optical strength for this class of materials which, due to their special structure and thermoelastic properties, differs greatly from that for conventional materials. Evaluation of the damage threshold of opaque polymers can be quite useful in laser-assisted etching processes.

Surface morphological studies, visual observation of plasma emission from the target, and reflectivity variation studies from the target are some of the methods utilized to evaluate the laser-induced damage threshold of materials. Techniques based on the photoacoustic effect have been shown to be very effective in determining the laser damage thresholds of both transparent and opaque samples (Rosencwaig and Willis 1980). Recently Petzoldt *et al* (1988) measured the damage threshold for bulk MgF_2, CaF_2 and LiF_2 by the photoacoustic deflection technique. In this paper we describe the use of the transverse photothermal deflection technique for the first time to evaluate the damage threshold of three bulk polymeric materials (perspex, nylon and Teflon) for 1.06 μm Nd–YAG laser radiation. The results indicate that the dependence of the photothermal signal (PTD) signal on laser energy for perspex samples has somewhat different behaviour than those of nylon and Teflon.

2. The photothermal deflection process

Absorption of laser radiation (pump beam) by a sample surface generates heat due to various non-radiative de-excitation processes occurring in the sample. The heat thus generated is transferred to the surrounding medium in the close vicinity of the irradiated surface, resulting in a temperature rise in the surrounding medium. Such a rise in temperature leads to density variations which create a refractive index gradient in the medium adjacent to the surface. A probe beam propagating through this refractive index gradient perpendicular to the direction of the pump beam will suffer refraction and consequently will deviate from its original path (Boccara *et al* 1980) corresponding to the ambient condition. This effect is termed PTD and, because of the geometry of the probe beam with reference to the pump beam, it is called

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transverse photothermal deflection. The magnitude of the beam deflection depends on the amount of heat transferred from the sample to the medium as well as the geometry of the heated area and the probe beam interaction. Heat transfer depends strongly on the thermal processes induced on the surface by the laser beam and it generally increases for pump energies above optical breakdown. As a result, at laser fluences at and above the damage threshold, a noticeable enhancement in PTD can be expected. Detailed theoretical analysis of optical beam propagation in an inhomogeneous medium with special reference to PTD has been given by Tam (1986) and Rose *et al* (1986). Since the probe beam profile is generally Gaussian, the extent of beam deflection can be measured using a position-sensitive detector (PSD) provided that the magnitude of the deflection is not too large.

To compute the thermal energy of a heated region by processing a detector signal, one must correlate the temperature distribution of the investigated region with both the optical beam propagation through the adjacent non-homogeneous medium and the detector response. The theoretical calculation of the probe beam deflection has been verified using a PSD such as a quadrant detector (Jackson *et al* 1980). In the present investigation, the polished tip of an optical fibre coupled to an avalanche photodiode (APD) acts as the position-sensitive detector (Rajasree *et al* 1990, 1992).

3. Experimental technique

A schematic diagram of the experimental set-up for determination of the damage threshold of polymer samples using the PTD technique is given in figure 1.

The sample, in the shape of a disc of 2.5 cm diameter

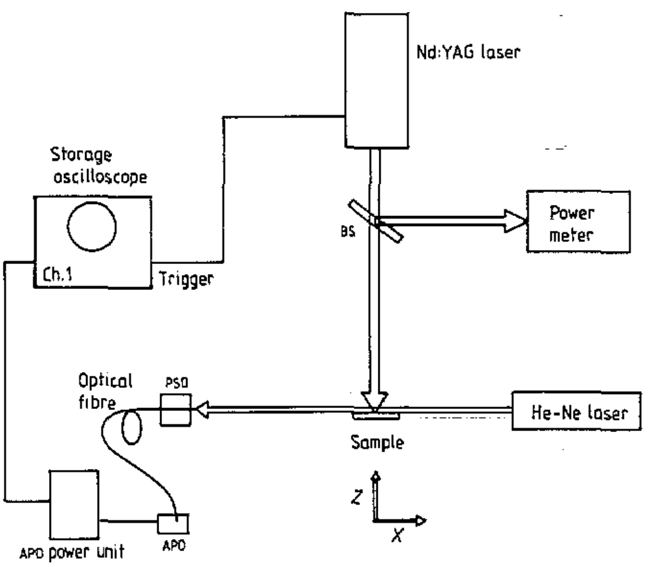

Figure 1. Schematic diagram of the experimental set-up: BS, beam splitter; PSD, position-sensitive detector; APD, avalanche photodiode.

and 0.55 cm thickness, is mounted on a micropositional XYZ translator. The pump beam used to irradiate the surface is pulsed 1.06 μm radiation (pulse duration 10 ns, single shot) from a Q-switched Nd–YAG laser (Quanta ray DCR11). A convex lens of short focal length ($f=$ 15 cm) focuses the pump laser beam on to the sample surface with more or less uniform intensity over the spot size. The laser fluence incident on the sample surface can be varied by adjusting the position of the lens in front of the sample. A stable 5 mW He–Ne laser beam (FWHM 0.8 mm) passing parallel to and grazing the sample surface is used as the probe beam. A fibre optical sensor, which acts as the position-sensitive detector, located about 50 cm from the sample measures the magnitude of the probe beam deflection. A 100 MHz storage oscilloscope (Tektronix 466) coupled to the PSD records this transient deflection which is a measure of signal amplitude. The pump laser energy is monitored for each pulse using a pulsed energy meter (Delta Developments) triggered in synchronization with the laser pulse. The sample was moved after each pulse and the signal amplitude is taken as the average value obtained at different fresh locations on the sample surface.

4. Results and discussion

Typical oscilloscope traces of the PTD signal recorded for nylon at different incident laser energy densities are shown in figure 2. The traces obtained for other samples also show the same qualitative features. The peak-to-peak value of the signal is taken as the signal amplitude.

Figures 3, 4 and 5 show plots of measured signal amplitude versus energy density of the pump beam incident on the sample for perspex, Teflon, and nylon respectively. These graphs exhibit distinct regions of different slopes, corresponding to different mechanisms for laser beam interaction with the surface of the material. The present investigations reveal an abrupt change in signal amplitude for regions near the threshold, which is in close agreement with the observations of earlier workers in similar materials. They have explained these regions in terms of surface damage, bond breaking and ablation (Milam 1977, Harada *et al* 1989, Rosencwaig and Willis 1980, Srinivasan and Bodil Braren 1989, Ravi Kumar *et al* 1991).

Even though the exact mechanisms of laser-induced damage in polymeric materials are not well understood, it has been observed that the laser damage threshold at A is found to be very sensitive to sample surface conditions. The value of the threshold at A is found to vary at different points on the sample surface for the three samples with an error of about 20%, whereas for region B the error is only 5%. The dependence of the damage threshold on possible absorptive inclusions, impurities and surface polishing of the sample has already been reported (Goldberg *et al* 1983, Srinivasan and Bodil Braren 1989). These include absorption by inclusions

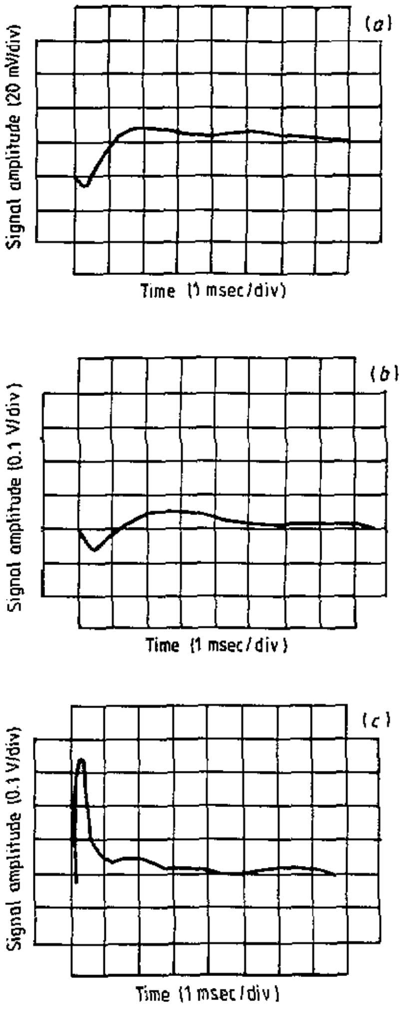

Figure 2. Oscilloscope trace of the signal from Teflon at: (*a*) 1.884 mJ cm^{-2} (20 mV/div; 1 ms/div); (*b*) 2.3 mJ cm^{-2} (0.1 V/div; 1 ms/div); and (*c*) 3.35 mJ cm^{-2} (0.1 V/div; 1 ms/div).

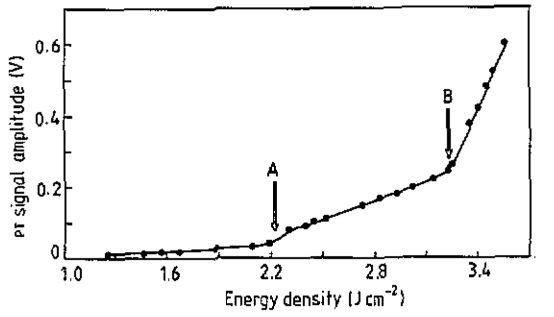

Figure 4. Plot of laser energy density versus PT amplitude for Teflon in air.

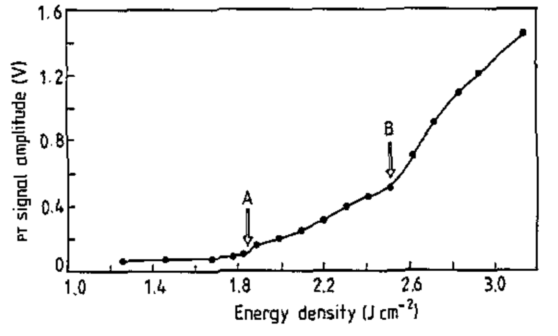

Figure 5. Plot of laser energy density versus PT amplitude for nylon in air.

and impurities, which can cause local heating and hence damage of the region irradiated by the high-power laser beam. Thus, of the two distinct regions of the curve, the threshold at A can be attributed to surface inhomogeneities. Below the threshold, almost the entire energy of the absorbed photons will be converted into heat, which in turn directly contributes to the photothermal signal. Hence, there is no significant change in the slope below

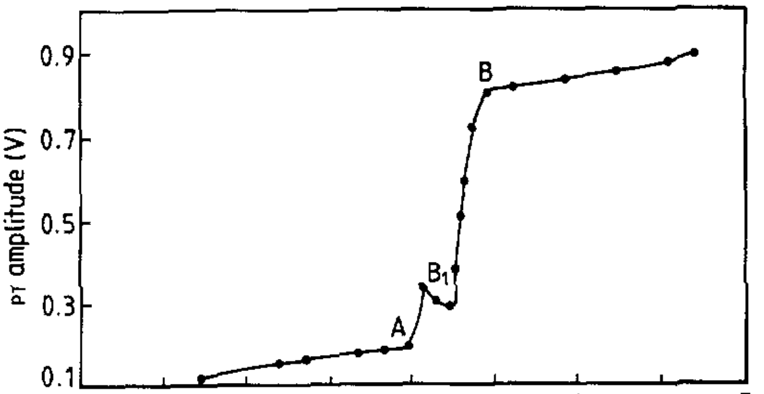

Figure 3. Plot of laser energy density versus PT amplitude for perspex in air.

the damage threshold. At A there is a noticeable change in the slope, which is an indication of the occurrence of damage to the surface of the material. The occurrence of surface damage upsets the thermal balance process in which, in addition to the surface layer, ambient air and the bulk of the sample are also involved. It is obvious that exceeding the damage threshold alters the thermal characteristics of the surface which controls the refractive index gradient above it. Under these conditions there is a greater amount of thermal diffusion (relative to conduction and convection of heat) (Shannon *et al* 1991) into the air adjacent to the surface and correspondingly an enhancement in PTD signal takes place.

In region B_1, the photothermal signal obtained in perspex shows a relative decrease while in polymer samples of nylon and Teflon such a decrease is imperceptible in the PTD. It is known that the signal amplitude decreases during an endothermic phase transition such as melting or vaporization (Rosencwaig and Willis 1980). Therefore some of the photon energy at this point is used up for the phase transformation, with a consequent reduction in PTD amplitude. This endothermic phenomenon is predominant for perspex in comparison with nylon and Teflon, as is evident from the plots. The optical penetration depth for transparent perspex is fairly large in comparison with those of nylon and Teflon and hence melting of the bulk of the sample is involved for perspex, whereas for nylon and Teflon only surface layers are involved in the optical absorption phenomenon. Correspondingly, the damage threshold is also higher for perspex. Well above the thresholds, further signal enhancement is evidently due to increase in nonlinear processes like multiphoton absorption. The second damage occurring at higher fluence (B) can be assigned to the mechanism of initiation of bond breaking and subsequent ablation of the material as a result of the increased multiphoton absorption (Rosencwaig and Willis 1980). It may be noted that the signal shape changes its polarity at the region B in figures 3, 4 and 5, corresponding to the ablation threshold. The shape of the signal now becomes similar to that shown in figure 2(*c*) which exhibits a prominent positive-going peak. The ablation products together with the vaporized material from the sample have a transient focusing effect, which could result in a positive-going part in the signal. Such changes of signal shape in PTD at the ablation threshold have been observed by others (Sell *et al* 1989). For samples that undergo a sharp increase in optical absorption coinciding with the damage, ablation and so on, the threshold can be readily determined by the sudden increase in the PTD signal.

The damage and ablation threshold values obtained for perspex, nylon and Teflon using the PTD technique are given in table 1. For comparison, the results obtained with alternative methods (Ravi Kumar *et al* 1991) are also included here and these values show good agreement with the results obtained from the present PTD measurements.

Table 1. The energy densities at regions A and B for perspex, nylon and Teflon. Estimated error about 20%. The alternative method data are from Ravi Kumar *et al* (1991)* and Milam (1977)**

Sample	Region A (J cm^{-2}) Present method	Region A (J cm^{-2}) Alternative method	Region B (J cm^{-2}) Present method	Region B (J cm^{-2}) Alternative method
Nylon	1.8	1.53*	2.5	2.25*
Teflon	2.2	1.78*	3.25	2.85*
Perspex	3.0	—	3.45	—
PMMA	—	1.6**	—	—
Polysterene	—	0.8**	—	—

5. Conclusion

The PTD studies provide with fair accuracy the magnitude of the threshold energy density required for laser-induced damage of target surfaces. These studies also give information about the different types of mechanism involved in laser-induced damage at different laser power densities. This technique offers an additional advantage over the photoacoustic method because it is more sensitive, since it avoids the acoustic impedance mismatch due to mechanical as well as acoustic coupling between the transducer and the sample needed in the latter technique. However, such detection techniques will work satisfactorily only in materials in which a significant increase in optical absorption accompanies laser damage.

Acknowledgments

The authors are grateful to the Council for Scientific and Industrial Research, Department of Science and Technology (Government of India) and the Ministry of Human Resources and Development for financial assistance. One of the authors (KR) is grateful to the University Grants Commission, New Delhi for a Junior Research Fellowship.

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