ELECTRON DENSITY DETERMINATION OF LASER INDUCED PLASMA FROM POLYMETHYL METHACRYLATE USING PHASESHIFT DETECTION TECHNIQUE

GEETHA K. VARIER, S. S. HARILAL, C. V. BINDHU, RIJU C. ISSAC, V. P. N. NAMPOORI and C. P. G. VALLABHAN
Laser Division, International School of photonics, Cochin University of Science & Technology, Cochin - 682 018, India

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Irradiation of a Polymethyl methacrylate target using a pulsed Nd-YAG laser causes plasma formation in the vicinity of the target. The refractive index gradient due to the presence of the plasma is probed using phase-shift detection technique. The phase-shift technique is a simple but sensitive technique for the determination of laser ablation threshold of solids. The number density of laser generated plasma above the ablation threshold from Polymethyl methacrylate is calculated as a function of laser fluence. The number density varies from $2 \times 10^{16} \text{ cm}^{-3}$ to $2 \times 10^{17} \text{ cm}^{-3}$ in the fluence interval $2.8-13 \text{ J cm}^{-2}$.

1. Introduction

The interaction of intense laser pulses with solid materials is of considerable current interest in view of the development of new techniques and devices involving non-linear effects. For example, laser ablation of polymeric materials is being used in photolithography and in the fabrication of micro electronic systems. Some of the transparent polymers are being used as optical components as well as non-linear optical materials. Very good optical transparency of Polymethyl methacrylate (PMMA) in the visible range of the electromagnetic spectrum helps them to be used as polymeric hosts for dye molecules which can be used in tunable solid state dye lasers. Also PMMA is very much suitable for the fabrication of optical fibre core as well as cladding. When pulsed laser radiation falls on the surface of an organic material, the surface layer is spontaneously etched away and the resultant molecular fragments get rapidly ablated from the target surface and the photo-chemistry is more or less simplified as explosive thermal decomposition. Materials used in high power applications should have high damage threshold under laser

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irradiation. The use of dye doped PMMA matrices in high power dye lasers demands the accurate knowledge of its laser damage threshold at different doping concentrations. Photothermal spectroscopy is a very sensitive method for studying laser matter interactions. In the present paper we illustrate the photothermal phase shift (PTPS) technique for the measurement of the laser ablation threshold of PMMA and also the number density of the resulting laser produced plasma when irradiated with a pulsed high power Nd:YAG laser beam.

2. Experimental

The experimental setup is as shown in Fig. 1. The basic element in PTPS technique is a Michelson Interferometer. Laser radiation from an intensity stabilized 5 mW He–Ne laser source (Spectra Physics) is used to construct the Michelson Interferometer (MI). Optical setup is aligned so as to get well defined straight fringe pattern. The beam in one of the arms of MI passes parallel and very close to the target surface. High power laser radiation from a pulsed Nd:YAG (Quanta ray DCR 11) laser at wavelength 1.06 μm with pulse duration 10 ns is focused on to the target in order to produce plasma.

The sample chosen for our study is a disc of PMMA having diameter 15 mm and thickness 4 mm. The point of irradiation is shifted by mechanically rotating the target after each measurement so that fresh location is available on the target surface for each pulse. The probe beam passes grazing to the sample surface so that the length of the plasma near the target is taken as equal to the pump laser spot size.
The shift in fringe pattern is measured as a voltage change using a PIN photo diode (HP-4207) and it is displayed on a digital storage oscilloscope (Jwatsu, 200 MHz). The whole setup has been properly vibration isolated by using an indigenously built vibration isolated table. Measurements were taken for different laser pulse repetition frequencies.

3. Results and Discussion

The presence of the plasma on one of the arms of the interferometer changes the effective path length on that arm and the corresponding phase change \( \gamma(x,t) \) will shift the fringe pattern which is proportional to the change in refractive index. This shift in fringe pattern can be measured as a voltage change \( \delta V(x,t) \) in the output of a photodiode given by,

\[
\delta V(x,t) \propto 2E_1E_2 \sin \left( \frac{4\pi}{\lambda} \int_0^1 \mu(x,t) dx \right)
\]  

(1)

Here \( l \) is the lateral extension of the plasma, \( \mu(x,t) \) the change in the refractive index due to the presence of the plasma, \( E_1 \) and \( E_2 \) are the amplitudes of the two interfering beams and \( \lambda \) the wavelength of the probe laser beam. For the intensity to be very sensitive to small changes in \( \gamma(x,t) \), the operating point is chosen such that the difference between the phases of the two interfering beams in the absence of the plasma is an odd multiple of \( \pi/2 \) i.e. \( \phi(E_1) - \phi(E_2) = (m + \frac{1}{2}) \pi \). Also when \( \phi(E_1) - \phi(E_2) \) is taken through a phase change of \( \pi \), \( (I_{\text{max}} - I_{\text{min}}) \propto 4E_1E_2 \) in the absence of plasma. Here \( I_{\text{max}} \) and \( I_{\text{min}} \) are the intensities corresponding to the bright and dark fringe centres respectively. Assuming a linear response for the photodetector, the corresponding voltage difference \( V_{\text{max}} - V_{\text{min}} \) denoted by \( V \) also is proportional to \( 4E_1E_2 \). The phase change \( \gamma(x,t) \) can be calculated from the measured values of \( V \) and \( \delta V \) and using Eq. (1) as

\[
\gamma(x,t) = \sin^{-1} \left( \frac{25V}{V} \right)
\]  

(2)

Using Eqs. (1) and (3) and \( \mu^2 = 1 - n_e/n_c \) one can arrive at the expression for the line averaged electron density

\[
n_e = \frac{k}{\lambda l} \gamma(x,t)
\]  

(3)

where \( k = 1.778 \times 10^{12} \text{ cm}^{-1} \). The line averaged electron densities were calculated using Eq. (3) for different laser fluences.

Variation of electron density with laser fluence (Fig. 2 pulse repetition rate 2.5 Hz) shows that electron density varies nonlinearly with respect to laser fluence. The graph exhibits regions of different slopes corresponding to different mechanisms for laser beam interaction with the target surface. At the point A there is an
abrupt change in the electron density of the plasma. It can be explained in terms of surface damage. For the values of the energy densities near the point $A$, the surface temperature is so high so as to produce intense ionisation, thereby causing rapid ionic and electronic emission from the surface. This rapid ionisation causes an abrupt change in the electron density which marks the surface damage threshold of PMMA at the laser fluence of $\approx 3.5 \text{ J} \cdot \text{cm}^{-2}$. Above the damage threshold there is a marked increase in the plasma electron density for a fluence up to about

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**Fig. 2.** Electron density of laser produced plasma as a function of laser fluence.

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**Fig. 3.** Variation of damage threshold with laser pulse repetition frequency. The solid line gives a linear fit to the observed data.
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$\approx 7.5 \text{ J} \cdot \text{cm}^{-2}$. Above this fluence the slow increase in electron density can be attributed partly to the increased electron-ion recombination forming a quasistable state and partly to the absorption of the incident laser radiation by the plasma plume.\(^12\) Figure 3 shows the dependence of the damage threshold on the pulse repetition frequency. At 16 Hz repetition frequency the ablation threshold comes down to 2.8 J $\cdot$ cm$^{-2}$ compared to 3.5 J $\cdot$ cm$^{-2}$ at 2.5 Hz. That is, the damage threshold decreases linearly with increasing laser pulse repetition frequency. At higher laser pulse repetition rate, damage threshold is low due to the influence of the thermal energy contribution from the previous pulse. This indicates that the process for ablation is thermal in nature.

The method of determination of electron density using interferometric technique has some advantages compared to other methods like spectroscopic methods and electronic probing.\(^13,14\) Spectroscopic determination of electron density requires absolute calibration of the detector in the entire spectral range of interest and in electronic probing, the flow pattern of the plasma is perturbed due to probe insertion. PTPS technique offers a simple, sensitive technique for determining the laser plasma densities and avoids the use of complex detection systems and painstaking calibration procedures.

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