CHAPTER - IX
LASER INDUCED DAMAGES

9.1 Introduction

Dye lasers offer a series of interesting spectroscopic properties, such as large spectral coverage and very small band width, which make them essential tools in a number of applications [1]. Recently a number of laser dyes have been described with the generation of stimulated radiation due to relatively novel photochemical mechanism [2-4]. Perylene [5-8] and p-terphenyl [9-11] dyes have exhibited lasing action and may be employed in the design of dye lasers. Optically induced damage to the laser material and to components of system generally determine the limit of useful performance of high power solid state lasers [12]. Now-a-days laser induced damage studies in thin films have received much attention [13-16], since an understanding of the damage mechanisms and a knowledge of the damage threshold of the film material are of great importance in designing a laser system in thin film form.

In the present work, perylene and p-terphenyl films prepared by vacuum deposition and ion plating techniques have been subjected to laser induced damage studies for the estimation of threshold energy densities.

9.2 Damage Mechanisms

9.2.1 General concepts

Laser induced damage can be defined as the physical appearance of permanently induced defect in the film surface of the material or as the onset of a degradation in the output performance of the laser system. Since there is no
direct relation of an observed damage centre with the degradation in laser performance, it is generally agreed upon to define the damage threshold as the energy or power density at which a change in physical appearance of the test component has been observed.

Damage may occur either internally or at the surface of the optical component. Internal damage is caused by particulate inclusions, small inhomogenities, absorption or self focusing in the material. Surface damage arises mainly from impurities, imperfections, inclusions or irregularities in the surface of transparent dielectrics [17]. Of the two, surface damage is generally more serious because it occurs at smaller values of incident beam energy.

The weakest link in a well designed laser is the surface of the optical components which serve as mostly reflective or antireflective coatings to reduce transfer losses. The survivability of thin film coatings to laser beam is of great importance in the reliable and efficient operation of high power laser systems. Due to surface imperfections and contamination, it is the surface of materials which generally fails first [18]. Substantial improvement in damage thresholds of optical surfaces has been achieved by empirical methods.

In order to develop quality coatings, one needs to understand the fundamental damage processes and their dependences on frequency and pulse length [19]. The three most frequently proposed processes of laser induced damage mechanism occurring in thin film material are avalanche ionization, multiphoton ionization and absorption by impurities within the films. Avalanche ionization occurs when an electron in the conduction band of a material absorbs enough energy from the electromagnetic field to impact-ionize a valence electron. The two electrons can
then undergo the same process to produce four conduction electrons and so on. The density of electrons in the conduction band grows exponentially with time until an absorbing plasma is formed. The energy efficiently absorbed by the plasma from the electromagnetic field leads to the occurrence of catastrophic damage in the material from locally deposited energy [20]. Multiphoton ionization can also produce a plasma; however in this case the electrons are promoted from the valence to the conduction band by direct absorption of two or more photons resulting in a plasma. The number of photons required for each ionization depends on the band gap of the material and the laser wavelength. Impurity dominated damage is a thermal process and can take place when an impurity in the film absorbs enough laser radiation to produce melting or fracture of the host material.

Extensive research has been carried out involving laser induced damage studies as a function of laser wavelength [21-25] pulse length [26-29] or film thickness [30-35]. Avalanche and multiphoton ionization theories predict conflicting laser wavelength dependencies, while the theories of impurity dominated damage predict little or no wavelength dependence. However, if the Mie absorption coefficient [20] is included in an impurity model, there is no longer a definite distinction between the wavelength prediction of the multiphoton and impurity model. This lack of distinction arises because the films can only be tested at a few discrete available wavelengths across the frequency spectrum. Furthermore, both theories predict an inverse dependence of damage threshold on laser frequency [20]. In general, studies of optical breakdown as a function of only one parameter cannot be expected to uncover the dominant mechanism of laser induced damage.
9.2.2 Impurity dominated damage

Sustained interest in the area of fundamental mechanism of laser induced damage is centered on describing the impurity dominated damage process in real optical materials [36,37]. In the case of impurity model, it requires a detailed knowledge of the type, size and distribution of the impurities in the host material. Because of the difficulties in evaluating these parameters, it is impossible to calculate the damage threshold by each of the competing theoretical approaches. Therefore, the only practical way to determine the most appropriate model is to examine their predictive ability in describing the variation of damage with easily controlled experimental or material variables such as pulse width, wavelength and film properties.

The role of the impurities has been considered as initiators in pulsed radiation. There has been a great deal of theoretical work on damage produced by impurities in dielectric host materials. Even now it is not clear whether the damage in very pure bulk material is produced by avalanche or multiphoton ionization, as opposed to isolated impurity sites. The impurity dominated breakdown mechanism assumes importance in the case of surfaces and thin films. This is due to the large absorption coefficients of thin films which ranges from 10 cm$^{-1}$ to 1000 cm$^{-1}$. For the same material in bulk form, the range is several orders of magnitude smaller. Approximately, surface absorption values fall between these two extremes.

The high absorptivity of thin films is generally attributed to impurities and imperfections which are included during the deposition process or infiltrate during exposure to the environment. Experimental data reveals that the damage threshold can be more than an order of magnitude lower for thin films than for the same material in bulk form.
In the impurity dominated damage model a spherical absorbing particle embedded in a host material is considered. This impurity absorbs the incident radiation and its temperature rises, which ultimately produces melting, vapourisation or stress fracture of the film material around the impurity. For dielectric impurities, in the form of a sphere an exact solution [38] to the thermal equation is given by

\[
T = \frac{3QI}{4\pi k_a} \left[ \frac{k_p}{3k_h} + \frac{1}{6} (1 - \frac{r^2}{a^2}) \right]
\]

\[
- \frac{2ab}{\pi} \int_0^\infty \exp \left( \frac{-y^2 t_p}{\gamma_1} \right) \frac{(\sin y - y \cos y) \sin (ry/a)}{y^2 (c \sin y - y \cos y)^2 + b^2 y^2 \sin^2 y} dy
\]

where

\[
Q = \text{absorption cross section} \quad r = \text{radial distance from the impurity boundary}
\]

\[
I = \text{incident laser intensity} \quad t_p = \text{laser pulse length}
\]

\[
a = \text{impurity radius} \quad k_p = \text{thermal conductivity of impurity}
\]

\[
k_h = \text{thermal conductivity of the host material} \quad D_p = \text{thermal diffusivity of impurity.}
\]

\[
D_h = \text{thermal diffusivity of the host material.}
\]

\[
\gamma_1 = \frac{a^2}{D_p} \quad b = \frac{k_h}{k_p} \sqrt{\frac{D_p}{D_h}}
\]

and

\[
c = 1 - \left( \frac{k_h}{k_p} \right).
\]
In the case of dielectric impurities it has been shown that the value of $Q^{[39, 40]}$ is given by

$$Q = \pi a^2 \left[ 1 + \frac{2 \exp \left( \frac{-8\pi n'a}{\lambda} \right)}{\frac{8\pi n'a}{\lambda}} + \frac{\{\exp \left( \frac{-8\pi n'a}{\lambda} \right) - 1\}}{\left( \frac{8\pi n'a}{\lambda} \right)^2} \right]$$

(9.2)

where $n'$ = imaginary refractive index and $\lambda$ = wavelength of light. It is seen that the cross section has dependence on wavelength and radius of impurity.

The most noticeable feature unique to the impurity model is the prediction of an increase in the damage threshold with a decrease in film thickness. This ensues from the quite reasonable assumption that the maximum size of the impurity is limited by the film thickness i.e., as the film thickness increases so does the impurity size. This observation is well documented by earlier works [20] in which this has been confirmed for many types of optical thin films.

Determination of the thermal properties of the inclusion and host is a difficult problem. Hence it will be worthwhile if a theoretical study of the sensitivity of the damage threshold to these quantities is made. Towards this end, Lange et al [41] have developed an approximate method of evaluating the integral in equation 9.1 by assuming that $[D_{tp}/a^2] > 1$. The radius at which damage first occurs is then given by

$$a_o = \frac{\sqrt{\pi D_h t_p}}{2}$$

(9.3)

and the damage threshold is

$$E_o = \frac{16T \sqrt{D_h c_h k_h t_p}}{\pi}$$

(9.4)
where \( c_h \) = specific heat density of the host material. The expression (9.4) not only verifies the \( \sqrt{\tau_p} \) dependence as suggested by Walker [20] but also predicts that the damage threshold scales linearly as the temperature at which damage occurs and as the square root of the product of specific heat and thermal conductivity of the host. In this approximation, the damage threshold is independent of the properties of the impurity.

9.3 Experimental

Generally, most of the damage thresholds reported so far have been obtained with lasers operated in the \( \text{TEM}_{oo} \) mode. Though the filamentary hot spots in the beam have higher energy densities compared to the average energy density, recent observations suggest that these filamentary structures are focused to spot sizes too small to contribute to damage. This implies that a \( \text{TEM}_{oo} \) mode laser is not necessary in laser damage studies [42]. Moreover, lasers are not always operated in the \( \text{TEM}_{oo} \) mode.

Three stages are involved in the measurement of laser damage threshold. The first stage is to irradiate the sample at several flux levels, some of which induce damage. The second is to measure the absolute characteristics of the pulse and the third is to determine which shot in the sequence caused damage. It is essential that the spatial and temporal profile of the laser remains a constant during the entire operations since there is no way to determine whether an unexpected result obtained, is due to the fluctuation in the laser or an exceptional property of the damage sample.
Figure 9.1 is a schematic diagram of the damage experiment. The laser employed in this study was a Nd : glass laser Q-switched by Kodak 14015 dye in 1,2-dichloroethane, with a pulse width of 25 ns (FWHM) at 1062 nm. The beam structure was multimode and was approximately Gaussian in profile with a diameter of about 4 mm. The laser output was passed through a biconvex lens of 17 cm focal length, onto the thin film sample. The sample mounted on a rotating platform was oriented, so that the angle of incidence of laser beam was 56°, thereby avoiding the multiple beam interference in the glass substrate. The laser pulse width was monitored on a Tektronix (Model 466 DM 44) storage oscilloscope with a Hewlett Packard hP 4207 photo diode and the energy incident on the sample by a 1 inch Scientech disc calorimeter (Model 38-0101). Experimental arrangement for laser induced damage threshold measurement is displayed in Figure 9.2.

Throughout a particular set of experimental study, the laser output energy was kept constant by maintaining a constant voltage on the capacitor bank of the flash lamp discharge circuit. With the present set up absolute energy measurement was not possible for every damage attempt. However, an energy calibration was made before and after each set of damage tests. To start with, the distance between the sample and the lens was adjusted such that the shot impinging on the film damages it. Then the film sample was pushed in the direction indicated by the arrow (Figure 9.1), to enable the next shot to impinge on a site adjacent to the previous one. During the shift of the film position due to the movement of the film away from the focus of the lens the power density incident on the sample is decreased. The film would get just damaged at a particular energy density known as the damage threshold. Once the damage threshold is attained, with the subsequent shots there will be no damage at all. During the damage on thin films, emission of bright light sparks was observed.
FIG. 9.1 SCHEMATIC REPRESENTATION OF LASER-INDUCED DAMAGE THRESHOLD MEASUREMENT

1. Q-SWIITCHED Nd: GLASS LASER
2. CALORIMETER
3. CONVEX LENS
4. SAMPLE
5. PHOTO DIODE
6. OSCILLOSCOPE
FIG. 9-2  EXPERIMENTAL ARRANGEMENT FOR LASER-INDUCED DAMAGE THRESHOLD MEASUREMENT
The damage sites were observed in a metallurgical microscope (Carl Zeiss Jena EPY-type : 2) after the completion of the test run and threshold damage location was identified. Typical photographs of the damaged sites are depicted by Figure 9.3. [Magnification = Magnification by objective x Magnification by eye-piece x ratio of the size of positive to that of negative of the photographic film.]

\[ M = 4 \times 3.2 \times 4 = 51.2 \approx 50 \].

Once the shot which caused the threshold damage is identified, one can calculate the threshold energy for the film by knowing the area of the site upon which the laser beam incident from the geometrical considerations. By measuring the pulse width using CRO one can estimate the power density of the incident laser beam.

9.4 Results and Discussion

The threshold energy densities estimated for the various films at different thicknesses are presented in Table 9.1. It is evident from the table that the threshold energy density increases with decrease in thickness for all the films and also the ion plated films are found to be with higher damage thresholds than the vacuum deposited ones.

For dielectric films [43, 44], it has been reported that the threshold increases with absorptance and decreases with increase in the film thickness [20]. As the most noticeable feature, unique to the impurity model is the prediction of an increase in damage threshold with a decrease in film thickness, the mechanism followed in these films may be an impurity dominated one. In this model, as discussed earlier,
<table>
<thead>
<tr>
<th>Film material</th>
<th>Technique of deposition</th>
<th>Film thickness Å</th>
<th>Threshold energy density J/cm²</th>
</tr>
</thead>
<tbody>
<tr>
<td>Perylene</td>
<td>Vacuum deposition</td>
<td>1470</td>
<td>19.880</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2040</td>
<td>7.170</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2950</td>
<td>4.971</td>
</tr>
<tr>
<td></td>
<td>Ion plating</td>
<td>1500</td>
<td>25.420</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2010</td>
<td>10.030</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2950</td>
<td>6.398</td>
</tr>
<tr>
<td>p-Terphenyl</td>
<td>Vacuum deposition</td>
<td>1050</td>
<td>17.500</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2090</td>
<td>4.980</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2470</td>
<td>3.660</td>
</tr>
<tr>
<td></td>
<td>Ion plating</td>
<td>950</td>
<td>22.870</td>
</tr>
<tr>
<td></td>
<td></td>
<td>1520</td>
<td>11.210</td>
</tr>
<tr>
<td></td>
<td></td>
<td>2100</td>
<td>5.928</td>
</tr>
</tbody>
</table>
the embedded spherical particle in a host material, absorbs the incident radiation resulting in temperature rise and causing damage to the film around the impurity. Hence it is quite reasonable that the maximum size of an impurity is limited by the film thickness i.e., as the film thickness increases so does the impurity size. Therefore the threshold energy density increases with decrease in film thickness. Such an inverse dependence of damage threshold on film thickness has been already reported [15].

The higher damage thresholds of ion plated films than those of vacuum deposited films may be attributed to the improved adhesion of films during ion plating process. Similar trend of damage threshold dependence on adherence of the films has been observed earlier [14,45].
References


