CHAPTER IX
LASER DAMAGE STUDIES

9.1 Introduction

The development and applications of high-power lasers during the past few decades have generated profound interest in the behaviour of optical materials under intense illumination. The demand imposed on optical materials, when they are incorporated into laser systems at high power levels, represent a new class of problem for the manufactures of optical elements. II-VI semiconductors are increasingly used in optical materials and semiconductor like CdTe, ZnSe and GaAs are possessing properties that are favourable for non-linear optical applications [1]. Hence a knowledge of the damage threshold of semiconductors and their alloys in thin film form is of great importance in designing the thin film laser devices and to operate it without appreciable degradation performance.

Now-a-days increasing interest is seen in the fabrication of laser system and the study of laser damage in thin films [2-10]. Electron beam induced crystallization of amorphous thin films of Se$_{80}$Te$_{20}$ alloy was studied by Lakshimi and Raghavan [2] and it was found that the internal stress accompanying the formations of crystalline nuclei disappered at high temperatures . Also laser induced crystallization of amorphous thin films of Se$_{80}$Te$_{20}$ alloy was studied at different laser beam intensities and different time durations by Lakshimi and Raghavan [4]. In the present work, thermally evaporated Te and Se$_x$Te$_{1-x}$ thin films have been subjected to laser induced damage studies for the estimation of threshold damage densities.
9.2 Laser Damage Mechanisms

Laser damage threshold can be defined as the physical appearance of defect in the material or as a degradation in the output performance of the laser system. Since it is very difficult to relate an observed damage centre to the degradation in laser performance, damage threshold is defined as the energy or power density at which a change in physical appearance of the test component has been observed. Damage threshold is specified in terms of the electric field associated with laser pulse.

The three most frequently proposed processes by which laser induced damage occurs in a dielectric thin film are (i) avalanche ionization (ii) multi photon ionization and (iii) absorption by impurities within the films. A brief description of the above processes are given in the following section.

9.2.1. Avalanche ionization

According to this model, avalanche ionization occurs when an electron in the conduction band of the 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.

It has been well established [11] that when the conduction electron density reaches a value of the order of $10^{18}$/cm$^3$ the absorption of the residual laser becomes appreciable, leading to localized heating and subsequent irreversible damage to the optical material. Therefore, the appearance of $10^{18}$ conduction electrons /cm$^3$ is taken as the criterion for laser-induced damage.
9.2.2. Multi photon ionization

In the case of multi photon ionization, 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. This model is based on the postulate that, when the photon energy of the incident light was about one third of the band gap energy (E_g) of the solid multiphoton absorption could contribute significantly to the breakdown process. The analytical difficulties of treating multiphoton absorption in dielectrics is mainly due to the lack of detailed knowledge of the band structure. There are several treatments of photon absorption to all orders [12, 13] of which the most widely used is that of Keldysh [12]. His formula gives meaningful results for first order as well as higher order photon process.

According to Keldysh's theory, the dependence of critical energy per unit area on the pulse width t_p reveals that if E_g < h\omega, the breakdown process is independent of time, where E_g is the band gap energy of the solids and \omega is the angular frequency of photon. For very high order photon process, E_g >> \hbar\omega, and the process depends linearly on time. Also it can be shown that the multiphoton theory predicts a decrease in the breakdown field with increase in laser frequency.

9.2.3. Impurity dominated breakdown

This damage is a thermal process and takes place when an impurity in the film absorbs enough laser radiation to melt or to fracture the host material. There has been a great deal of theoretical work on damage produced by impurities in host materials [14,15]. The role of impurity dominated breakdown assumes importance in the case of surfaces and particularly in thin films. This is because of large absorption coefficient of most thin films which ranges from 10 cm\(^{-1}\) to 1000 cm\(^{-1}\). This high absorptivity of thin films is
generally attributed to impurities which are included during the deposition process or infiltrate during exposure to the environment.

The most noticeable feature unique to the impurity model is the prediction of an increase in damage threshold with a decrease in film thickness. This ensures from the quite reasonable assumptions 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.

9.2.4 Choice of the mechanism

Considerable amount of research has been carried out on laser induced damage studies as a function of laser wavelength [16 - 20], pulse width [21-24] or film thickness [25-30]. The frequency dependence of laser damage has frequently been proposed as the main theoretical bench mark in distinguishing between the competing processes [31]. This results mainly because, avalanche and multiphoton ionization theories predict conflicting laser wavelength dependence while theories of impurity dominated damage predict little wavelength dependence. However, if the Mie absorption coefficient [32] is included in an impurity model, there is no longer a definite distinction between the wavelength predictions of the multiphoton and impurity models. This lack of distinction arises because the films can only be tested at a few discrete available laser wavelengths across the frequency spectrum. Further more, both theories predict an inverse dependence of damage threshold on laser frequency [33]. In general, studies of laser induced breakdown as a function of only one parameter cannot be expected to uncover the dominant mechanism involved.
9.3. Measurements

Three stages are involved in the measurement of laser damage threshold density, viz., (i) irradiation of the sample at several flux levels (some of the levels will induce damage) (ii) measurement of absolute characteristics of the pulse and (iii) identification of the shot in the sequence which cause damage.

Fig. 9.1. shows the schematic diagram of the experimental set up to measure the damage threshold. DCR-11 quanta ray (Q-switched mode) Nd: YAG laser is used, which emits pulse width of 1 ns duration at 1.06 μm wavelength. The laser output of beam diameter 3mm is passed through a biconvex lens of focal length 0.19m onto the thin film samples. The samples are mounted in such a way that they are exposed to the laser beam normally. The energy at the thin film sample is determined by a pulsed laser energy meter (Delta Developments, England).

The distance between the sample and the lens is kept constant and the laser output is varied until the shot impinging on the film damages it. Then the thin film sample is pushed to new position to enable the next shot to impinge on a site adjacent to the previous one. The particular energy at which the film gets just damaged is taken as damage threshold and it is identified by observing sparks of bright light emission or hearing sound from the sample area. Once the damage threshold is attained, with the subsequent shots, there will not be anymore damage at all. The energy density is calculated by knowing the area of the damaged site from the geometrical considerations. Throughout the study, the laser is operated in TEM$_{01}$ mode. Mostly, the other reported damage thresholds have been obtained by the laser operated in the TEM$_{00}$ mode. Though the filamentary hot spots in the beam have higher energy density, recent observations suggest that these filamentary structures are focussed to spot sizes too small to contribute to the damage. This implies that a TEM$_{00}$ mode laser is not necessary in laser damage studies [33].
FIG. 9.1. SCHEMATIC DIAGRAM OF EXPERIMENTAL SETUP FOR LASER DAMAGE MEASUREMENT.

1. Nd:YAG Laser
2. Mirror
3. Lense
4. Beam splitter
5. Sample
6. Energy meter
After attaining threshold, a number of damage sites are examined with the help of a metallurgical microscope (Versamet -2 union 7596, Japan).

9.4 Results and discussion

9.4.1. Threshold damage energy

The laser induced damage threshold densities estimated for Te and Se$_x$Te$_{1-x}$ films of different compositions (Se$_{0.7}$Te$_{0.3}$, Se$_{0.5}$Te$_{0.5}$ and Se$_{0.3}$Te$_{0.7}$) are presented in Tables 9.1 to 9.4. It is clear from the Table 9.4 that the threshold energy density decreases with increase in thickness of Se$_{0.3}$Te$_{0.7}$ films. Earlier investigators have reported that the threshold energy density increases with absorptance [6, 9 - 11] and decreases with increase in film thickness of dielectric films [32,34,35].

Surface damage in thin films arises mainly from impurities, imperfections, inclusions or irregularities on the surfaces [36]. Even though the parent material is pure, impurities and imperfections are included in thin films during the atomistic condensation process or infiltration during exposure to the environment. The infiltration and absorption of oxide impurities in vacuum deposited germanium films have been observed by Kameswara Rao et al [37], who studied the effect of GeO amorphous phase in the germanium films exposed to atmospheric air, and the growth of GeO crystalline phase on the same sample on subsequent exposure to laser beam. In addition to the infiltration of oxygen, the absorption of other impurities from the atmospheric air into the film cannot be ruled out, since the laser damage study is being carried out in normal atmospheric air. These facts and the decreasing nature of damage threshold with increase of the film thickness indicate that the mechanisms followed in these film materials are impurity dominated ones. In this model, a spherical absorbing particle embedded in the material is considered. This impurity absorbs energy from the incident radiation and its temperature rises, which causes melting, vapourization or stress fracture of the
film material around the impurity. This model proposed for dielectric films also holds good to explain the results of the present study.

A reverse trend has been noticed for (Te, Se$_{0.7}$Te$_{0.3}$, and Se$_{0.5}$Te$_{0.5}$) films (Table 9.1 to 9.3), i.e., the threshold energy density increases with the increase in film thickness. According to Russel Austin et al [38], among the various parameters the inherent film stress also a major variable for studying the laser damage threshold in thin films. The increase in damage threshold with increase in film thickness for (Te, Se$_{0.7}$Te$_{0.3}$, and Se$_{0.5}$Te$_{0.5}$) films may be explained on the basis of stress present in the films. Many workers reported that the film stress decreases with increase in film thickness [38,39]. Similar behaviour has been observed for Te, Se$_{0.7}$Te$_{0.3}$, and Se$_{0.5}$Te$_{0.5}$ films in the present investigation. As the film thickness increases, the homogeneity of the film increases which in turn reduces the stress present in the film. The increase in laser damage threshold with increase in film thickness for Te, Se$_{0.7}$Te$_{0.3}$, and Se$_{0.5}$Te$_{0.5}$ films may be due to the decrease in stress at higher thicknesses. The increase in threshold with thickness have been already reported for (ITO) [6] and polymer films [4].

### 9.4.2 Laser damage patterns

Optical materials in the thin film form are the most damage sensitive elements in high power laser systems. Hence the resistance of thin film coatings to laser beams is of great importance for the reliable and efficient operation of high power laser system. When the laser beam passes through the film, according to the energy of the pulses, different patterns were observed in the films.

Damage spots having different shapes and sizes indicate the total damage at those sites. Fig 9.2(a,b) shows the optical micrographs of damage spots in a tellurium film of thickness 176 nm. Laser damage initiates single
hole defects. These single holes coalesce to create total damage to the film. Initiation of single holes in Te film is shown in Fig 9.2 (a). These single holes merge together with the increase of the energy of the pulses resulting in the total destruction of the film [Fig 9.2(b)]. FIGS. 9.3 to 9.5(a) shows the optical micrographs of laser damage sites of Se$_{0.7}$Te$_{0.3}$, Se$_{0.5}$Te$_{0.5}$ and Se$_{0.3}$Te$_{0.7}$ thin films of thicknesses 129, 110 and 138 nm respectively. These three micrographs clearly shows the single hole propagating nature in the three different compositions. The complete damage pattern in the form of circular rings is shown in FIGS. 9.3 to 9.5(b) Se$_{0.7}$Te$_{0.3}$, Se$_{0.5}$Te$_{0.5}$ and Se$_{0.3}$Te$_{0.7}$ thin films. The damages are confined to a circular area.
FIG 9.2 (a,b) SINGLE HOLE PROPAGATING AND COMPLETE DAMAGE SITE MICROGRAPHS OF Te THIN FILMS
FIG. 9.3 (a, b) SINGLE HOLE PROPAGATING AND COMPLETE DAMAGE SITE MICROGRAPHS OF Se$_{0.7}$Te$_{0.3}$ THIN FILMS
FIG. 9.4 (a,b) SINGLE HOLE PROPAGATING AND COMPLETE DAMAGE SITE MICROGRAPHS OF $\text{Se}_{0.5}\text{Te}_{0.5}$ THIN FILMS
FIG. 9.5 (a,b) SINGLE HOLE PROPAGATING AND COMPLETE DAMAGE SITE MICROGRAPHS OF $\text{Se}_{0.3}\text{Te}_{0.7}$ Te THIN FILMS
Table 9.1
Material (Te)

<table>
<thead>
<tr>
<th>Film thickness (nm)</th>
<th>Threshold energy density (J/cm²)</th>
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<tbody>
<tr>
<td>76</td>
<td>60.5</td>
</tr>
<tr>
<td>134</td>
<td>77.0</td>
</tr>
<tr>
<td>197</td>
<td>110.0</td>
</tr>
<tr>
<td>295</td>
<td>195.0</td>
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Table 9.2
Material (Se₀.₇Te₀.₃)

<table>
<thead>
<tr>
<th>Film thickness (nm)</th>
<th>Threshold energy density (J/cm²)</th>
</tr>
</thead>
<tbody>
<tr>
<td>92</td>
<td>0.15</td>
</tr>
<tr>
<td>155</td>
<td>0.196</td>
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<tr>
<td>190</td>
<td>0.223</td>
</tr>
<tr>
<td>243</td>
<td>0.420</td>
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Table 9.3
Material (Se_{0.5}Te_{0.5})

<table>
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<th>Film thickness (nm)</th>
<th>Threshold energy density (J/cm²)</th>
</tr>
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<tr>
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<tr>
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<td>169</td>
<td>0.410</td>
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<td>213</td>
<td>0.920</td>
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</table>

Table 9.4
Material (Se_{0.3}Te_{0.7})

<table>
<thead>
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<th>Film thickness (nm)</th>
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<tr>
<td>79</td>
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<tr>
<td>138</td>
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<tr>
<td>183</td>
<td>39.4</td>
</tr>
<tr>
<td>205</td>
<td>34.5</td>
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