IOSR Journal of Applied Physics (IOSR-JAP)
e-ISSN: 2278-4861.Volume 9, Issue 5 Ver. I (Sep. - Oct. 2017), PP 37-44
www.iosrjournals.org

Influence Of Laser Irradiation On The Optical Properties Of In_6Se_7 Thin Film Synthesized By Thermal Evaporation Technique

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***Abstract**: Thin films of In_6Se_7 were prepared by thermal evaporation technique under a vacuum of 10^{-4} Pa then exposed to laser irradiation from diode laser HPL532-Q10W. The effects of laser-irradiation on the optical parameters were investigated. The optical parameters were acquired utilizing spectrophotometric measurements of the transmittance and reflectance at normal incidence of light at the wavelengths range 200–2500 nm. The type of optical transition was found to be an indirect allowed transition for pristine film and laser irradiation films. It was found that the optical band gap diminishes by increasing the time of laser-irradiation. On the other hand, the dispersion parameters of In_6Se_7 films were decreased by increasing the irradiation time.*

Keywords: Chalcogenide glasses; thermal evaporation; thin film; laser irradiation effect; Optical properties

Date of Submission: 16-08-2017 Date of acceptance: 05-09-2017

I. Introduction

Chalcogenide compounds are sensitive to external influences, especially laser-irradiation, heat treatment because of their flexible structure. According to the previous studies, the role of laser irradiation is to create some microstructural changes in film and bulk materials [1]. Chalcogenide semiconductors have shifting of band gaps and are transparent in the IR region. Moreover, they are interesting as core materials for optical fibers used for light transmission, especially when short lengths and flexibility are required. Layered compounds form a class of materials in which a significant energy density can be stored in the form of mixed ionic electronic conductors [2]. Indium Selenide is an important material of III–VI group compounds. The energy gap of InSe at room temperature is 1.3 eV, which makes it an attractive material for solar energy conversion, infrared device, lasers and diodes [3,4]. It is likewise used as a promising material for optoelectronic devices, heterojunction devices. However, little work has been reported concerning the thin film state [5]. Although there have been several studies on the growth and characterization of In_6Se_7 thin films, deposited by different growth techniques, there still a lack of understanding on the optical properties which strongly affect device performance, All the reported works on InSe thin films lack information about the dispersive optical parameters like the oscillator strength, oscillator energy, static dielectric constant and static refractive index .and also lack some external effects such as the effect of laser irradiation on optical properties of In_6Se_7. The present study deals with an experimental investigation of the effect of films thickness and time of laser irradiation on the optical properties of thermally evaporated In_6Se_7 thin films.

The transmittance and reflectance measurements were carried out at room temperature over the wave length range from 200 to 2500 nm. As a result of optical energy gap studies, the optical energy band gap was found to be between 0.92ev and 1.3 ev, depending on time of laser irradiation. The objective of this work is to produce In_6Se_7 thin films by using thermally evaporated deposition technique and to investigate the influence of laser-irradiation and effect of thickness on their optical properties.

II. Experimental Details

In_6Se_7 compound was synthesized first, by mixing and melting the high purity (99.999%) individual elements in stoichiometric proportions in a sealed quartz ampoule [6] The resultant bulk was then used as the source material for the deposition of In_6Se_7thin films [7]. In_6Se_7thin films were prepared by thermal evaporation on to cleaned glass substrates for optical measurement at vacuum of 10^{-4} Pa using an Edward E306A coating unit the evaporation rates as well as the film thickness were controlled by using a quartz crystal monitor FTM4 [8]. The thickness of dissipated films was measured by a multiple beam interferometry method. Some of these films were irradiated with laser for different time range from 5 min to 20 min in air at room temperature.

The transmittance (*T*) and reflectance (*R*) of the as-deposited and irradiated films on glass substrates were determined at normal incidence in the wavelength range 200–2500 nm by means of a double-beam spectrophotometer (JASCO model V-570 UV–vis-NIR) to determine some optical parameters of as deposited In_6Se_7 films and study the effect of laser irradiation on these parameters in the same thicknesses, and also study the effect of thicknesses on these parameters. The absolute values of transmittance and reflectance were used to determine the optical constants such as refractive index (*n)*, and absorption index (*k)*, by applying a special computer program by using Mat lab [9,10]. based on Merman's exact equations.

III. Results And Discussion

The spectral behaviors of *(T %)* and (*R%)* for as-deposited and laser-irradiated In_6Se_7 thin films with a thickness of 70 nm, as a representative sample, are illustrated in Fig(1).

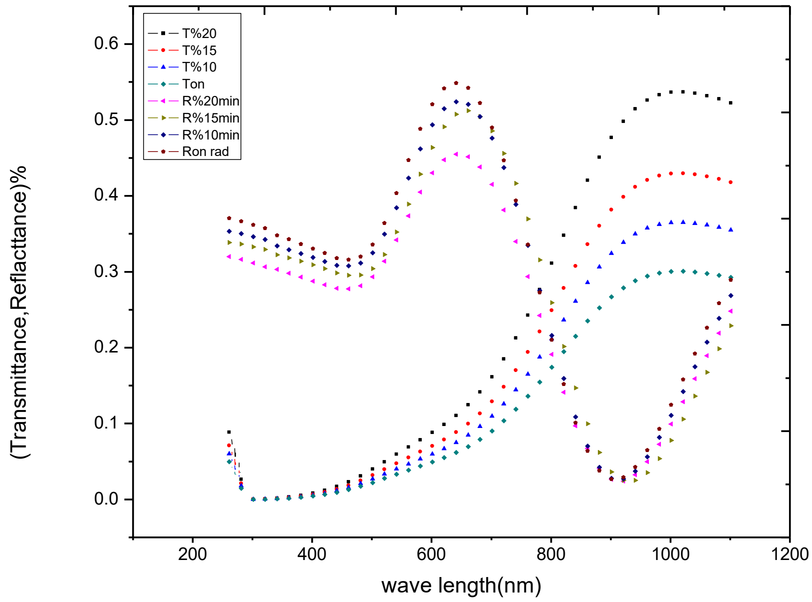

Fig-1-Spectral behavior of transmittance, *T*, and reflectance, *R*, for as-deposited and laser irradiate In_6Se_7 thin films

Transmittance is almost zero up to 500 nm and then started increasing with increasing wavelength. It suggests that this material can be used as blocking film for wavelength up to 500 nm [11]. It is also observed that the reflectance decrease with increasing the time of laser- irradiation. It is also observed that the transmittance edge shows a shift towards a higher wavelength scale showing that the energy gap will be diminished with increasing laser-irradiation time. Here also irradiation of minute's time interval produces harmonic effect in reflectance. Estimations of Transmittance and Reflectance with various laser irradiation times are given in Table (1).

Table (1): Transmittance (T %) and Reflectance (R%) of In_6Se_7 with and without laser irradiation at 70 nm

Sample (s)	Pristine sample	Irradiated sample		
		10minutes	15minutes	20minutes
T % At wave length(950nm)	0.3	0.36	0.44	0.55
R% At wave length(600nm)		10minutes	15minutes	20minutes
	0.55	0.52	0.5	0.46

Curves show that the transmittance have values (~0.3-0.55) at **λ** =950 nm and the reflectance have values (~0.46-0.55) at **λ** =600 nm. This behavior indicating that there is negligible absorption beyond the saturated transmittance values. It is also observed that the transmittance edge shows a shift towards a higher wavelength scale showing that the energy gap will be diminished with increasing laser-irradiation time.

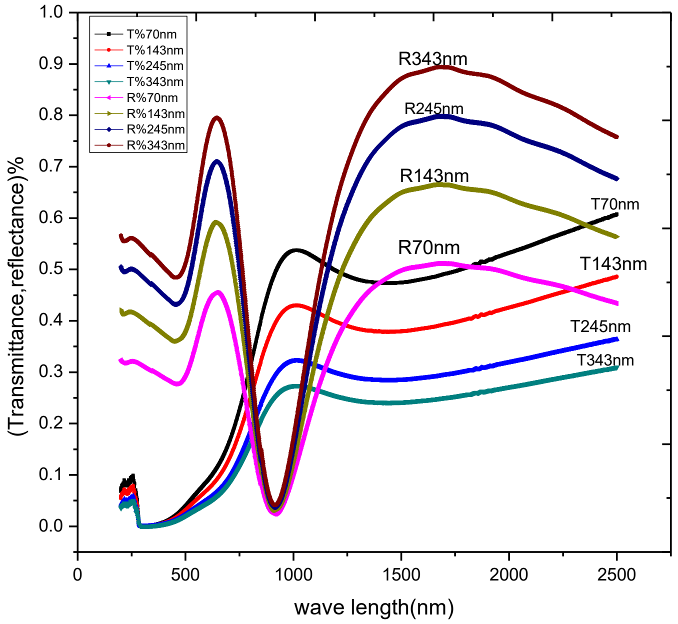

Fig-2-Spectral distribution of normal incidence transmittance, *T*, and reflectance, *R*, for In_6Se_7thin films with different thickness

The spectral distribution of both *T* and *R* of the as-deposited In_6Se_7 thin films with different thicknesses are depicted in Fig.2. The results clearly show that the transmittance decreases rapidly with the increases in film thickness, it may be due to the increase in opaqueness, indicating the cat ion vacancies [12], Transmittance continuously decreases with wave length from near infrared through visible region due to the increase of scattering loss at porous surface [13].
Absorption coefficient α can be calculated by using the equation [14]:

$$\alpha = \left(\frac{1}{d}\right)\ln\left(\frac{(1-R)^2}{T}\right) \qquad (1)$$

Where d is thickness of film in cm, (T, R) are Transmittance and Reflectance respectively
The relation between absorption coefficient (α) and photon energy (hν) is appeared in Figure (3).

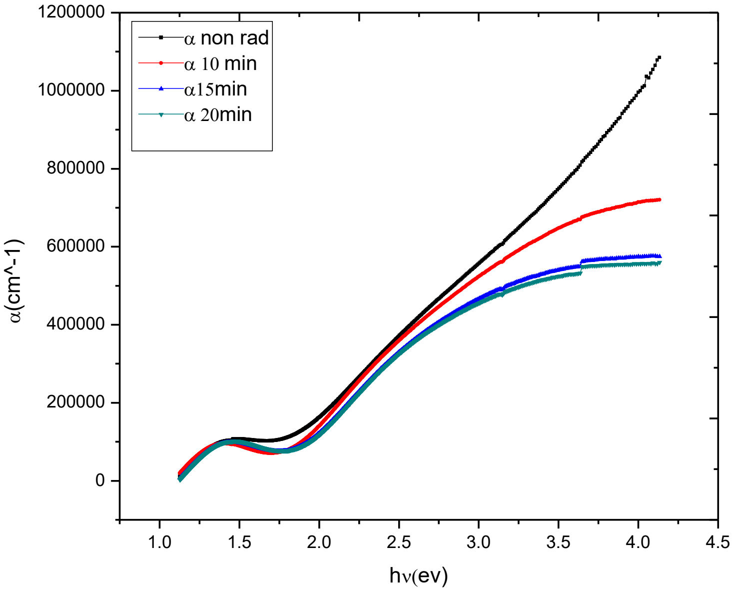

Fig-3-Variation of absorption coefficient with photon energy for $In_6Se_7$with and without laser irradiation

This is quite reasonable, since near the fundamental absorption edge, the absorption coefficient varies rapidly with energy. The electronic move between the valence and the conduction bands begins at the absorption edge corresponding to the minimum energy difference between the lowest energy of the conduction band and the highest energy of the valence band in the material. Also we can observed that the absorption coefficient decrease by increasing the time of laser- irradiation. Optical energy gap (Eg) can be calculated by following expression:

$$\alpha (h\nu) = A (h\nu - E_g)^m \qquad (2)$$

Where A is the constant, Eg is the optical energy gap of the material and m is a number which characterizes the transition process involved. m has the value (1/2) for the direct allowed transition and 2 for an indirect allowed transition. According to Tauc [15,16]the absorption tail related to localized states into the pseudo-gap, which Localized states can arise from the existence of vacancy defects and/or impurities. The optical band gap(E_g) have been measured from the plot $(\alpha h\nu)^{1/2}$ versus $h\nu$ by extrapolating the curves to $h\nu$ axis at $(\alpha h\nu)^{1/2} = 0$ .For all samples before and after laser irradiation with different time as shown in figure (4).

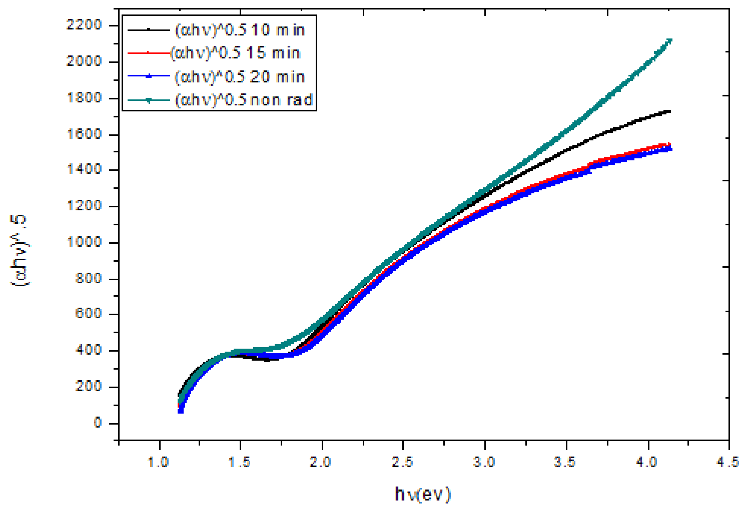

Fig-4-Variation of $(\alpha h\nu)^{1/2}$ versus $h\nu$ for $In_6Se_7$with and without laser irradiation

From this figure we can observed that the Variation of $(\alpha h\nu)^{1/2}$ versus $h\nu$ for $In_6Se_7$and with without laser irradiation give straight line that say the type of the transition for In_6Se_7 is indirect allowed transition [17]. from fig (4) we can determined the optical energy gap for In_6Se_7thin film with and without laser irradiation as shown in figure (5).

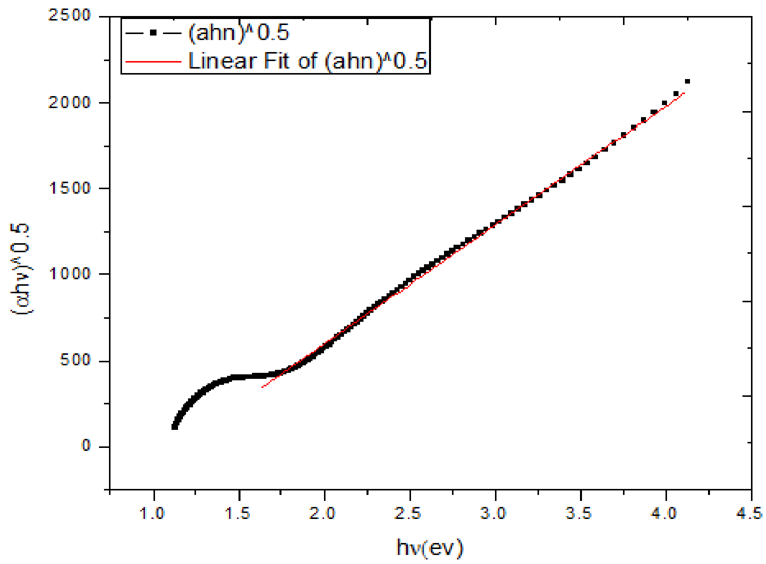

Fig-5-Optical E_g for In_6Se_7 thin film (70nm) as prepared

The calculated data shows that the optical energy gap decreases with increase laser irradiation time as shown in table (2).

Table (2): Optical E $_g$ for In_6Se_7 thin film (70nm) Pristine sample and laser irradiated.

Sample (s)	Pristine sample	Irradiated sample		
		10 minutes	15 minutes	20 minutes
Optical band gab	1.14ev	1.096ev	0.966ev	0.921ev

Valance band of chalcogenides forms by lone pair orbital whereas conduction band is formed by anti-bonding orbital. Laser irradiation excite the electron from the lone pair of bonding state to higher energy states and hence Vacancies created in these states are immediately filled by the outer electrons by Auger process that in turns induce more holes in the lone pair bonding orbitals leading to a vacancy cascade process which makes easier bond breaking and ionization of atoms and changes the local structure order of the amorphous network causing a decrease in the optical energy gap. Influence of laser irradiation on the optical properties is connected with higher degree of disorder in the alloy. Hence the increase in transition probability due to disorderness produced by laser irradiation leads to narrowing the optical band gap of In_6Se_7alloy [18].fig (6) show the relation between the optical energy gap and the time of laser irradiation.

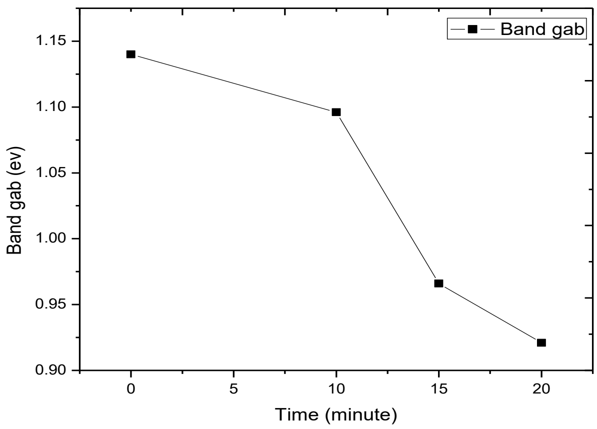

Fig-6-Variation of optical band gap with laser irradiation time in In_6Se_7

The optical properties of the solid are governed by the interaction between the solid and the electric field of the electromagnetic wave. The extinction coefficient k is related to the damping of the oscillation amplitude of the incident electric field. The extinction coefficient (k) is a measure of the damping factor, which indicates the amount of absorption loss when the electromagnetic wave propagates through the material, has been calculated using well known relation [19].

$$k = \frac{\alpha * \lambda}{4\pi} \qquad (3)$$

Variation of extinction coefficient with incident energy as well as with laser irradiation time is shown in figure (7) .

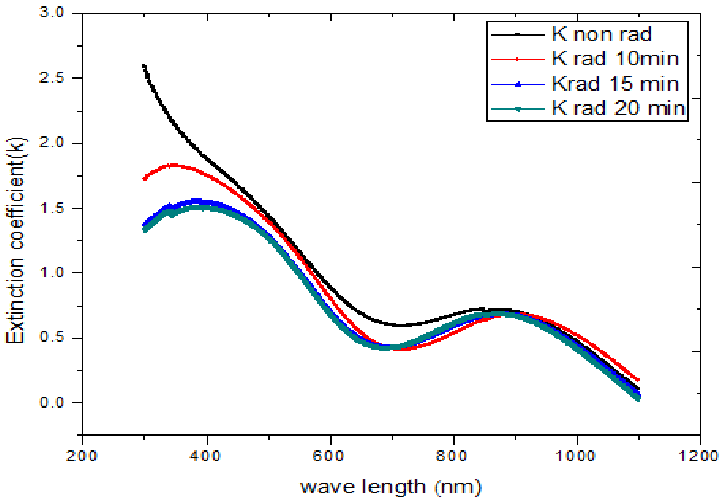

Fig-7-Variation of extinction coefficient (k) with wave length for different time of laser irradiation.

Extinction coefficient decreases with increasing the wave length of the incident light however laser irradiation causes harmonic pattern with remarkable overall decrease in extinction coefficient.
The refractive index (n) has been calculated using-

$$n \quad \frac{1+\sqrt{R}}{1-\sqrt{R}} \qquad (4)$$

Figure (8) show that the refractive index increase with increasing the time of laser irradiation.

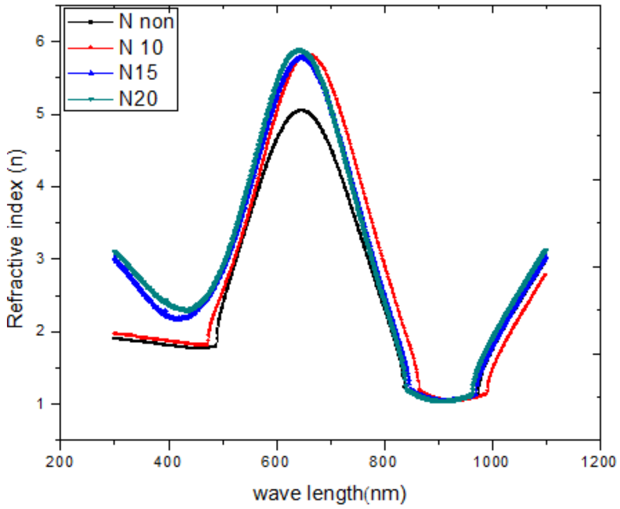

Fig-8-Variation of ε_i with photon energy for different time of laser irradiation

Also, the complex dielectric constant ε described the optical properties of material and calculated in the following equation:

$$\varepsilon \;=\; \varepsilon_r + \; i\varepsilon_i \qquad (5)$$

Where ε_r and ε_i represent the real and imaginary parts of dielectric constant respectively, and determined by the following equations

$$\varepsilon_r = \; n^2 - \; k^2 \qquad (6)$$

And,

$$\varepsilon_i \;= 2nk \qquad (7)$$

Variation of these constants with photon energy for different irradiation time has been plotted in Figures 9and 10 respectively.

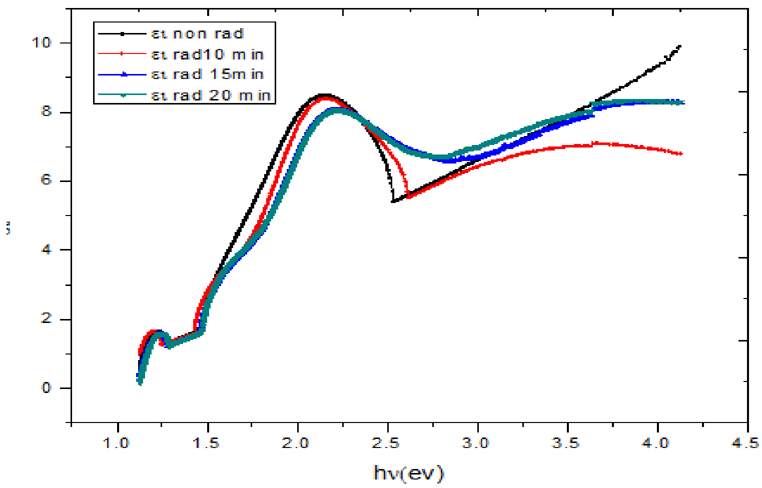

Fig-9-Variation of ε_i with photon energy for different time of laser irradiation.

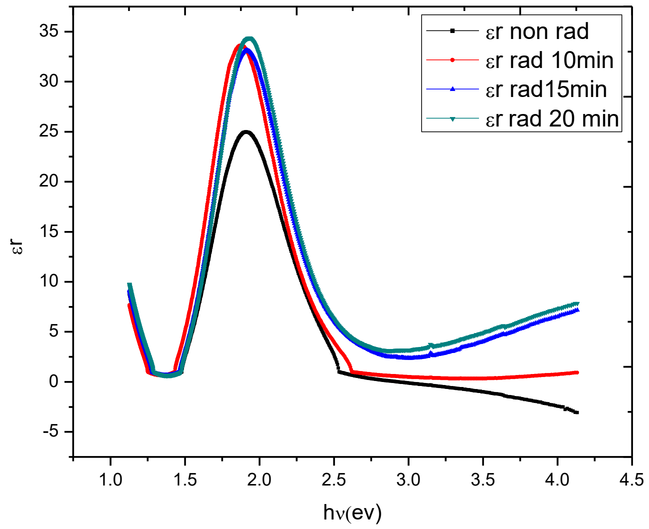

Fig-10-Variation of ε_r with photon energy for different time of laser irradiation.

IV. Conclusion

In this study In_6Se_7 thin films were prepared by thermal evaporation technique. The effect of laser irradiation on optical properties of In_6Se_7films in different irradiation times have been studied. The spectral behavior of transmittance, *T*, and reflectance, *R*, for such films recommends them to be good optical filter materials. The optical constants *n* and *k* are affected by film irradiation. The type of transition in as-deposited and irradiated films were indirect allowed with an optical energy gap range from (1.21 to 0.92ev) .The laser irradiation decreases the value of the energy gap. The laser irradiation a slight increase of the value of determined dispersion parameters such as dielectric constant. These constants are changing in a harmonic pattern which can be of great interest for laser activated switches and memories. These changes can be utilized as 0 and 1 stages in laser tuned digital memory devices fabrication.

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Fatma Gami. "Influence Of Laser Irradiation On The Optical Properties Of In6Se7 Thin Film Synthesized By Thermal Evaporation Technique." IOSR Journal of Applied Physics (IOSR-JAP), vol. 9, no. 5, 2017, pp. 37–44.