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M. M. Abd El-Raheem
Physics Department, Faculty of Sciences, Sohag University, Sohag 82524, Egypt, elneh@yahoo.com

H. E. Hassan
Cyclotron Facility, Nuclear Physics Department, Nuclear Research Center, Egyptian Atomic Energy Authority, Cairo13759, Egypt, elneh@yahoo.com

M. M. Wakkad
Physics Department, Faculty of Sciences, Sohag University, Sohag 82524, Egypt, elneh@yahoo.com

H. M. Ali
Physics Department, Faculty of Sciences, Sohag University, Sohag 82524, Egypt, elneh@yahoo.com

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The Effects of Gamma Irradiation on the Optical and Electrical Properties of Melt Quench Ge18Bi4Se78 Chalcogenide Glass.

M. M. Abd El-Raheem[1,], H. E. Hassan[2], M. M. Wakkad[1], H. M. Ali[1], A. K. Diab[1], S. K. Mohammed[1] and H. F. Mohamed[1]*

[1]Physics Department, Faculty of Sciences, Sohag University, Sohag 82524, Egypt
[2]Cyclotron Facility, Nuclear Physics Department, Nuclear Research Center, Egyptian Atomic Energy Authority, Cairo13759, Egypt

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Abstract: The structural, optical, and electrical properties of as-deposited and gamma irradiated (50, 100, 150kGy) Ge18Bi4Se78 thin films have been investigated. The structural characteristics of both the as-deposited and gamma irradiated films are inspected by X-ray diffraction (XRD).The optical constants of all the films are analyzed in the wavelength range 250-2500 nm employing spectrophotometer measurements at normal incidence. The type of transition is estimated using the obtained optical constants. The optical energy gap Eop as well as Urbach Eu energy in addition to plasma frequency ωp are studied. Single oscillator and Drude models are used to discuss the refractive index in the normal dispersion region. The effect of γ irradiation on the DC conductivity of the considered films is inspected.
Keywords: Ge18Bi4Se78; thin film; gamma irradiation; optical band gap; refractive index; dc conductivity

1 Introduction

The chalcogenide glass is used as an important component in a specific device; it must be fulfilling the condition of thermally saturated. This is because when such a glass is prepared, generally by melt quenching technique, it is formed in a non-equilibrium thermodynamic state. Therefore, it is possible that physical properties of a given glass may deteriorate upon usage at temperatures even below the glass transition temperature [1]. Besides, the glass may be annealed at different temperatures for different annealing times before its final usage. Whereas this process takes long period of time, It was reported that [2-4] It is preferred to use external influences such as gamma ray, accelerated electrons, neutrons, lasers, etc. to accelerate thermal saturation in order obtain the thermally relaxed glass proper for particular applications.

A change in the physical, chemical, mechanical and electrical properties of the semiconductor materials upon exposure to ionizing radiations such as x-rays, beta particles, gamma-rays and alpha particles. These changes can enhance the performance of various materials and create new materials. It has been reported that amorphous semiconductors can be used as radiation dosimeters for industrial applications [5-8]. The effects of high energetic ionizing gamma ray irradiation on chalcogenide glasses films have been investigated [9]. Irradiation changes the physical properties of the materials [10], these changes are substantially dependent on the internal structure of the absorbed materials. The irradiation might cause ionization or excitation for the electrons and may be displace the atoms from their lattice sites. The produced photoconduction electrons become loosely bounded to some trapping centers elsewhere in the material structure [3]. These new electronic array cause a change in the optical properties of the semiconducting films. Therefore, these changes in the optical properties of the chalcogenide glasses under influence of gamma irradiation make them attractive for variety of applications. Few studies have been reported on the influence of gamma irradiation on the optical properties of chalcogenide [11-13].

The fundamental experimental measurements are used to evaluate several structural and optical parameters such as the density, molar volume, excess volume, optical band gap, free volume percentage, the compactness, and packing factor. The results show that as the bismuth content increased from 0 to 12 at. %, the compactness, the optical band gap, and packing density decreased, whereas the density, molar volume, excess volume and free volume

*Corresponding author e-mail: elneh@yahoo.com

percentage increased [14]. The effect of the gamma irradiation exposure by different doses on the optical properties and dispersion parameters for chalcogenide glasses $Se_{70}S_{30}$-xSb_x thin films are investigated [15]. Accordingly, the refractive index of the investigated films increases with raising the doses of gamma irradiation, this can be interpreted in terms of the increase of the density of the investigated films with exposure to gamma irradiation due to atomic displacements or ionization. The calculated dispersion (strength) energy E_d, static refractive index n_o, zero frequency dielectric constant ε_o found to increase after irradiation while the single oscillator energy E_o, reduced after irradiation [15.]. Furthermore, increase of the doses of gamma radiation resulted in increasing the absorption coefficient. In contrast, the obtained optical energy gap of $Se_{70}S_{30}$-xSbx films was found to decrease with increasing the doses of gamma radiation which can be attributed to the increase of the defects after irradiation. This is assured by the decrease in the Urbach energy E_u after radiation.
Croitoru et.al [16] have shown a change of mechanism of conduction due to the damaged regions, and the irradiation process usually results in the creation of structural defects [17-18]. The average sizes of grains of the gamma irradiated samples found to be 4-5 times lower than that of the non-irradiated one [19].
Therefore, the aim of the present work is to investigate the effect of γ-irradiation the optical energy gap, Urbach tail, refractive index, dispersion parameters and plasma frequency of Ge_{18} Bi_4Se_{78} chalcogenide glasses thin films.

2 Methodologies

5N purity of Ge, Bi, and Se were used for preparing bulk amorphous samples of the system Ge_{18} Bi_4 Se_{78}. Appropriate mole percentages of the three elements were mixed and enclosed in 10 mm diameter silica tube then sealed off after evacuation of the order of $10{-}5$ torr and heated in an oven. The temperature was raised slowly to reach 1000ºC and kept for 30 hrs then quenched in ice water. Thin films of thickness 500 nm were deposited using Edwards high vacuum coating unit model E306 on ultrasonically clean glass substrate where the deposition rate was 10 nm/min. and the thickness was controlled using thickness monitor model TM 200 Maxtek. The substrate temperature was maintained at room temperature. Investigations of the microstructure were carried out using XRD model D8 Advanced Bruker. The optical transmittance T and reflectivity R were measured by means of a computer programmable Jasco V-570 (Japan) double beam spectrometer in the wavelength range from 200 to 2500 nm at normal incidence. For reflectivity measurements an attachment model ISN-470 was provided.
The resistivity measurements were achieved using two terminal designs by applying constant voltage to the sample and count the outcome current using Keithely 614 electrometer. Silver paste is used as electrodes contacts over the film surface of separation 2 mm.
To study the effect of γ-radiation on the prepared Ge_{18} Bi_4 Se78 thin film, three samples were exposed to ^{60}Co source at variable γ -ray doses of 50, 100 and 150 kGy. Irradiation was performed using gamma ray irradiator model CM-20 at the cyclotron facility of Egyptian Atomic Energy Authority. The certified dose rate was 0.5 kGy/h according to the previously reported data [20]. The sample temperature was monitored by a thermocouple attached to the sample surface during irradiation and it wasn't raised above 30 oC during the experimental time. Therefore, any enhanced effect on the properties was considered due to irradiation. The irradiated samples were characterized by the same techniques mentioned above and the results were compared with that of pristine thin film.

3 Results and Discussions

3.1 Structural Analysis

The XRD diffraction patterns of the as-deposited and gamma irradiated $Ge_{18}Bi_4Se_{78}$ thin films reveal the amorphous nature due to disappearance of any peak represent crystallization as shown in Fig. 1.

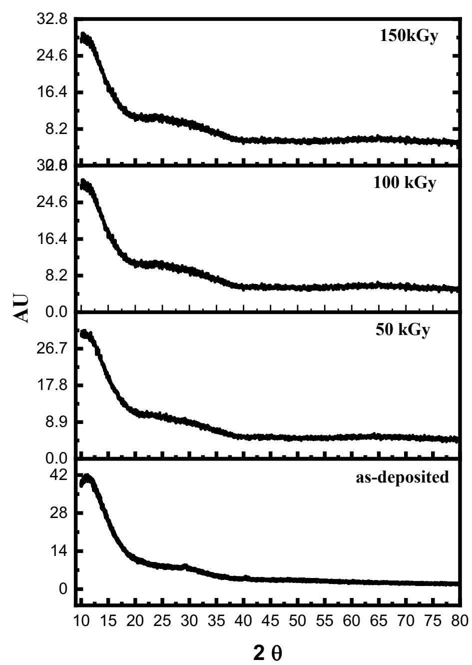

Fig. 1: XRD patterns of as-deposited and gamma irradiated Ge18BixSe78-x chalcogenide glass thin films.

3.2 Optical Analysis

The transmittance T(λ) and reflectance R(λ) spectra in the range of wavelength from 200 to 2500 nm are shown in Fig. 2. In this region, interference fringes are created due to back and forth bouncing off the incoming wave through interfaces air–film, film–substrate, and substrate–air. Occurrence of these fringes considered to be an evidence of the high quality and symmetry of the deposited films [20]. The number of interference fringes decreases with increasing γ irradiation dose.

The absolute values of the transmittance T(λ) and reflectance R(λ) are given by [21].;

$$T = \frac{I_f}{I_g}\,(1 - R_g) \qquad (1)$$

Since I_f and I_g are the intensities of light crossing through the film-glass system and that passing across the reference glass, respectively and R_g is the reflectance of the glass substrate, and

$$R = (\frac{I_r}{I_m})R_M(1 + [1 - R_g]^2) - T^2R_g \qquad (2)$$

where I_m is the intensity of light reflected from the reference mirror, If r is the intensity of light reflected from the sample and R_M is the mirror reflectance.

For calculating the optical constants, absorption coefficient, α, the extinction coefficient, k, of the films at various wave lengths, the following equations are used [22-23];

$$\alpha = \frac{1}{d}Ln[\frac{(1-R)^4}{4T^2} + \sqrt{\frac{(1-R)^4}{4T^2} + R^2} \qquad (3)$$

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

d is the thickness of the film equal 400 nm.

The refractive index n((λ) can be calculated using Swanepoel's approach as the following expression [24]:

$$n^2 = N_1 + \sqrt{N_1^{2-}N_2^2} \qquad (5)$$

$$N_1 = 2N_2(\frac{T_M - T_N}{T_M T_N}) + (\frac{N_2^2}{2}) \qquad (6)$$

T_M and T_m are the maximum and minimum values of transmittance at a particular wavelength, N2 is the refractive index of the substrate [25]

$$n = (\frac{1+R}{1-R}) + \sqrt{\frac{4R}{(1-R)^2} - k^2} \qquad (7)$$

α is the absorption coefficient and d is the film thickness. According to Konstantinov et.al [26], the experimental errors in the calculated values of k and n with accuracy better than ±4%, measuring the film thickness as ±2%, and in T and R as ±1% are taken in account.

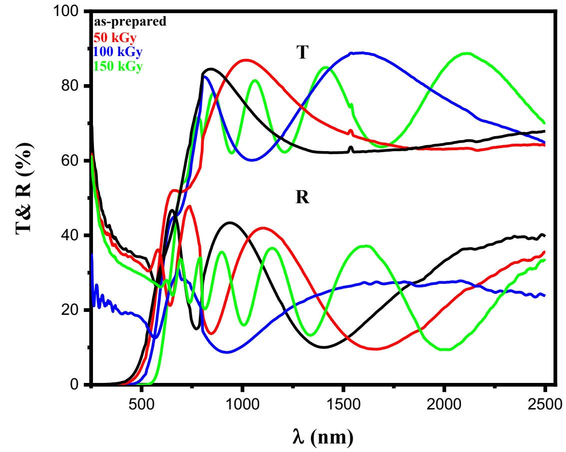

Fig.2: Transmittance and reflectance spectra of as-deposited and gamma irradiated $Ge_{18}Bi_xSe_{78-x}$ thin films.

This figure reveals that the transmittance and the reflectance curves are systematically affected by γ-irradiation. In the absorption region (400 nm-700 nm), the transmittance spectra of the irradiated films are shifted to shorter-wavelength (blue shift), and show larger transmittance than that of the as-deposited film. Such shifts are associated with change in the optical gap of the irradiated films. While in the transparent region (700 nm-2000 nm), it can be observed that the as-deposited film shows an increase of the transparency from 78% at 770 nm up to 89% at 2115 nm. The irradiated film at 50 kGy shows an increase from 44% at 640 nm up to 89% at 1600 nm. The irradiate film at 100 kGy shows an increase from 44% at 640 nm to 90% at 1600 nm, in addition, for irradiated film at 150 kGy the transmittance found to increase up to 85% at 850 nm then decreases with more prolonging of the wavelength.

The type of transition and the optical energy gap values can be explained by using the [19]:

$$(\alpha h\nu) = C(h\nu - E_{op})^p \qquad (8)$$

where C is a constant and p=1/2 and 3/2 for direct allowed and forbidden transitions, respectively, p = 2 and 3 for indirect allowed and forbidden transitions, respectively. The dependence of $(\alpha h\nu)^{1/p}$ on photon energy (hν) was plotted for different values of p. The best fit was obtained for p = 2 and illustrated in Fig.3; this result indicates that the transitions are indirect allowed transitions. The indirect band-gap determination is based on the extrapolated linear regression of the curve resulting from a plot of $(\alpha h\nu)^{1/2}$ versus photon energy ($h\nu$). The extrapolation of the straight line graphs $(\alpha h\nu)^{1/2}$ gives the values of the optical gap for the as-deposited and γ-irradiated films. The estimated values of the indirect optical gap show slight decrease from 1.84 eV for as-deposited film to 1.48 eV after γ-irradiated with γ-dose 150 kGy (redshift). Y. The variation of indirect optical gap with γ-dose is displayed in Fig.4.

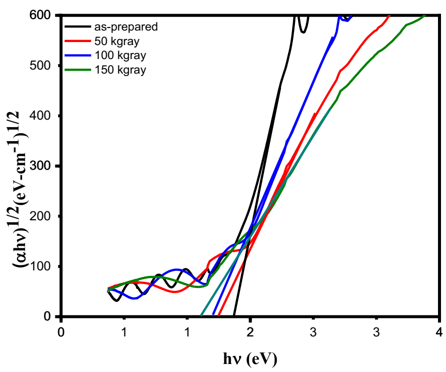

Fig.3: The relation between $(\alpha h\nu)^{1/2}$ and photon energy $(h\nu)$ of the as-prepared and gamma irradiated $Ge_{18}Bi_xSe_{78-x}$ thin films.

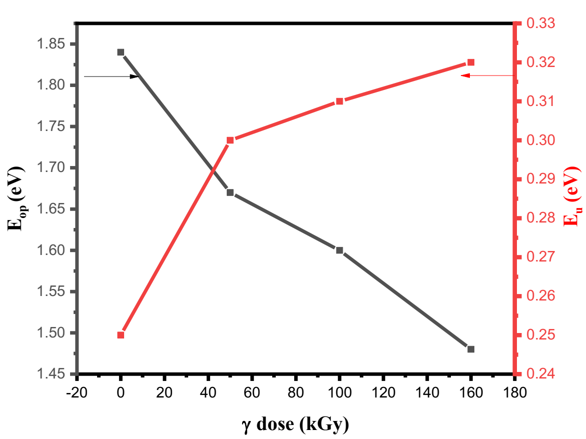

Fig.4: Variations of the optical energy gap and Urbach energy with gamma dose.

Sharma and Maity [27] reported that the optical band gap of $(TeO_2)_{0.9}$ $(In_2O_3)_{0.1}$ thin films was found to decrease from 2.7 to 2.5 eV after γ-irradiation with dose 75 Gy. This decrease in the optical band gap is due to the increase in Urbach tails. It is recognized that the oxygen vacancies are known as color centers. Zhu [28] supposed that ionizing radiation causes structural defects which intern leading to a change in their density upon exposure to γ-ray. The produced free electrons as a result of band to band transitions are trapped in oxygen ion vacancies which in turn is the cause of formation of color center leading to increase the electrical conductivity [29]. Therefore, the decrease in the optical band gap is essentially due to the increase in Urbach tails [30]. The defects are created in the film during γ-irradiation and get annihilated even under the normal room temperature status [31]. The creation and annihilation of defects are coexist together and at higher doses of γ-irradiation (50– 150 kGy), where the number of defects created as a result of γ-irradiation equals the number of annihilated defects [32]. The wideness of the localized states in the energy gap (Urbach tail) Eu can be calculated using Urbach equation [32]:

$$\alpha = \alpha_o e^{\frac{h\nu}{Eu}} \tag{9}$$

E_u is Urbach energy and α_0 is a constant. The value of E_u can be determined from the slope of the straight lines of the plot $\ln(\alpha)$ vs. photon energy $(h\nu)$ relation as seen in Fig.5. All the calculated values of E_{op} and E_u and are plotted as a function of γ dose in Fig.4 and recorded in Table 1. It is denoted that E_{op} decreases and E_u increases with increasing γ dose.

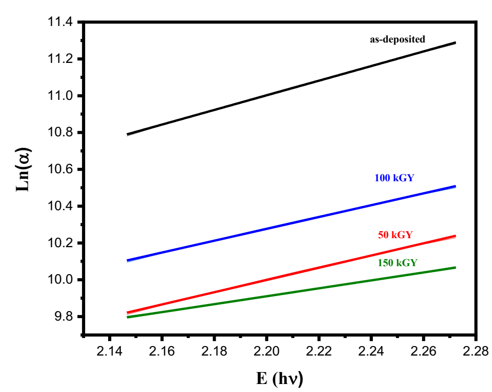

Fig.5: The relation between $\ln(\alpha)$ and photon energy $(h\nu)$ of the as-prepared and gamma irradiated $Ge_{18}Bi_xSe_{78-x}$ thin films.

Optical properties of $Ge_{18}Bi_4Se_{78}$ thin films can be described by refractive index n(λ) and extinction coefficient k(λ) that comprises the complex index of refraction. Fig. 6 shows the real part of the refractive index, n(λ), for as-deposited and γ-irradiated $Ge_{18}Bi_4Se_{78}$ thin films. It is observed that the refractive index has a behavior of normal dispersion over the wavelength range $\lambda > 600$ nm. At longer wavelength, 0.75 μm – 2.5 μm, the average calculated value of n decreases by an amount $\Delta n = 1.16$ after γ-irradiated with dose 150 kGy. Also, there are slight variations in the position and the highest intensity of the refractive index as a result of γ-irradiated.

Fig. 7 shows the spectral behavior of the absorption coefficient α for as-deposited and γ-irradiated $Ge_{18}Bi_4Se_{78}$ thin films. It can be seen that the intensity and the absorption edge of α are considerably decreased with increasing gamma dose.

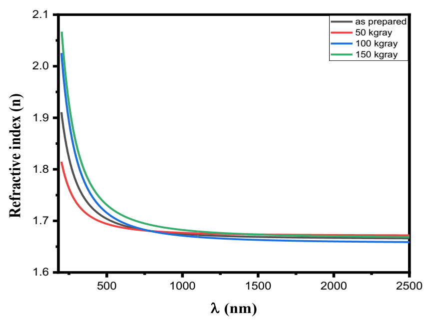

Fig.6: Refractive index spectra of the as-prepared and gamma irradiated $Ge_{18}Bi_xSe_{78-x}$ thin films.

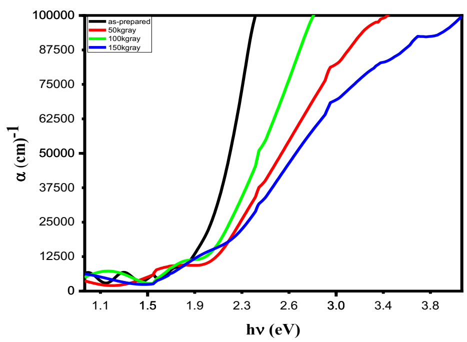

Fig.7: plots of absorption coefficient α with photon energy $h\nu$ for the as-prepared and gamma irradiated $Ge_{18}Bi_xSe_{78-x}$ thin films.

The variation of the refractive index n(λ) in the region of very small values of the extinction coefficient k(λ) under negligible damping is provided by the classic dispersion theory . Wemple and DiDomenico [33-34] described the wavelength dependence of the refractive index, n(λ), in the transparent region for various different solids by using the single-oscillator model of the form;

$$(n^2 - 1) = \frac{E_o E_d}{E_o^2 - (h\nu)^2} \tag{10}$$

where (hυ) represents the photon energy, E_o is the oscillator energy and E_d is the dispersion energy describes the strength of the electronic transitions. The calculated values of the dispersion parameters as well as the infinite frequency dielectric constant, ε_1, obtained by plotting of $(n^2\text{-}1)^{-1}$ versus $(h\nu)^2$ for the as-deposited and irradiated $Ge_{18}Bi_4Se_{78}$ thin films are shown in Fig. 8. Besides, the dispersion theory is used to obtain the lattice dielectric constant, ε_L via the plot of n^2 versus λ^2 .The relation between the real dielectric constant, ε_1, and λ^2 in the transparent region is given by [35];

$$\varepsilon_1 = n^2 = \varepsilon_L - \frac{e^2 N}{4\pi\varepsilon_o m c^2}\lambda^2 \tag{11}$$

where ε_L, e, ε_o, an N/m* are the lattice dielectric constant, the elementary charge, is the permittivity of free space and is the ratio of free carrier concentration to the free carrier effective mass respectively. Fig. 9 shows the variations of n^2 with λ^2 for the as-deposited and irradiated $Ge_{18}Bi_4Se_{78}$ thin films. It is observed that the dependence of n^2 on λ^2 is linear at longer wavelengths. Extrapolating the linear parts to zero wavelength gives the value of ε_L and from the slopes of the linear parts the free carrier concentration, N, is obtained assuming that the optical effective mass of the electrons in $Ge_{18}Bi_4Se_{78}$ is about $m^*=0.35\ m_o$ [36], where $m_o = 9.11\times10^{-31}$ kg. The variations of ε_L and ε_1 with γ-irradiation dose are shown in Fig. 11. It is obvious from this figure that ε_L and ε_∞ increasing with increasing irradiation dose, while the disagreement between ε_L and ε_1 may be due to free carriers' contribution [24]. According to Drude model, the plasma frequency (ωp) is related to the free charge carrier density, N, by the relation [37]

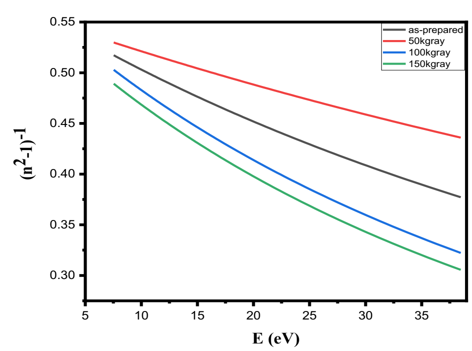

Fig.8: The changing of $(n^2\text{-}1)^{-1}$ with $(h\nu)^2$ for the as-prepared and gamma irradiated $Ge_{18}Bi_xSe_{78-x}$ thin films

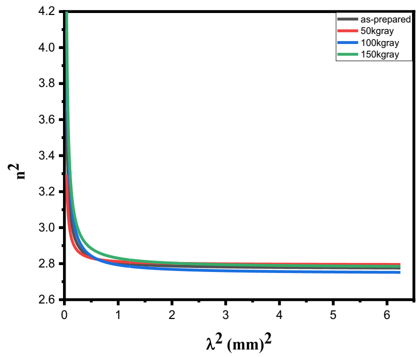

Fig.9 : The changing of n^2 with λ^2 for the as-prepared and gamma irradiated $Ge_{18}Bi_xSe_{78-x}$ thin films.

Table 1: Values of the optical energy gap E_{op}(eV), Urbach energy E_u(eV), single oscillator energy E_o(meV) , dispersion energy $E(meV)_d$, ratio of carrier concentration to effective mass N/m x10^{54} (cm-^{3}g^{-1}), ω_px10^{15} (sec^{-1}), lattice dielectric constant, infinity dielectric constant at different values of gamma dose (kGy).

γ dose	E_{op}	E_u	E_o	E_d	N/m^*	ω_p	ε_L	ε_∞
0.0	1.84	0.25	1.99	2.16	1.28	1.02	2.94	1.09
50	1.67	0.30	2.24	3.07	1.88	1.24	3.66	1.37
100	1.60	0.31	2.8	4.95	1.97	1.27	3.70	2.17
150	1.48	0.32	2.34	5.18	2.35	1.38	4.4	2.21

3.3 Electrical Analysis

The variations of the current density J with applied field is demonstrated in Fig.10 revealing Ohmic behavior representing semi-metal state. Also, it is evident that J increases with increasing gamma dose.

The changing of the dc electrical conductivity with gamma dose is demonstrated in Fig.11 revealing increasing the conductivity in the range of dose from 0 to 50 kGy. This can be interpreted as follows; the resultant free electrons due to band to band transitions are trapped in defect centers leading to the increase of the electrical conductivity [29]. By increasing the dose of gamma from 50 kGy to 150 kGy; the conductivity decreases, this behavior agree well with the reported researches [16-17].

The results reported by Croitoru et.al [16] have shown a change of mechanism of conduction due to the damaged regions, where localized levels are created, which are the main cause of the deviation of the electrical characteristics of the non-irradiated films from those irradiated ones. Also, irradiation process usually results in the creation of structural defects; the recovery effect of irradiation is also recognizing [17]. The average sizes of grains of the gamma irradiated samples found to be 4-5 times lower than that of the non-irradiated one [18].

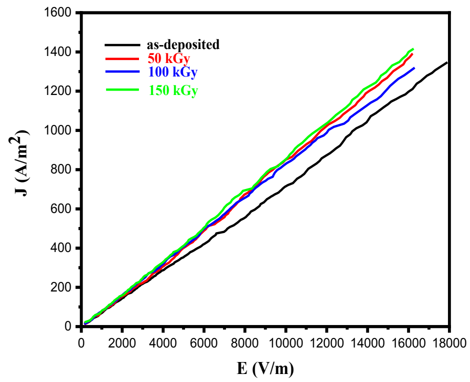

Fig.10: The relation between the current density J with applied electric field E for the as-prepared and gamma irradiated Ge18BixSe78-x thin films.

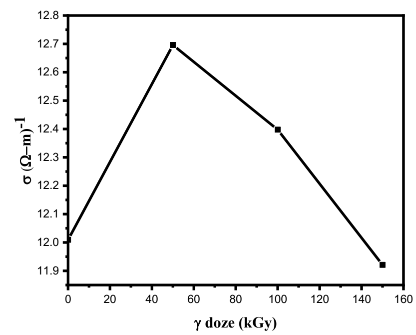

Fig.11: Changing of dc conductivity σ with gamma dose γ.

4 Conclusions

Thermal evaporation method is used to deposit thin films from melt quenched $Ge_{18}Bi_4Se_{78}$ chalcogenide glasses. XRD analysis revealed the amorphous nature of the considered thin films. Optical energy gap of the $Ge_{18}Bi_4Se_{78}$ chalcogenide glasses found to decrease with increasing the gamma ray dose; in contrast, Urbach energy decreases. The refractive index behaves as a normal dispersion and found to increase with increasing gamma dose. The obtained plasma frequency, lattice dielectric constant, infinitely dielectric in addition to the dispersion energies E_o and E_d found to be increase with increasing gamma dose.

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Conflict of interest: The authors declare that there is no conflict regarding the publication of this paper.

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