DIYALA JOURNAL FOR PURE SCIENCES

Effect of irradiation on the optical properties of PbSe
thin film prepared by thermal evaporation method
Jasim Mohammed Abid Al-Leteef

Effect of irradiation on the optical properties of PbSe thin film prepared by thermal evaporation method

Jasim Mohammed Abid Al-Leteef

University of Diayla   College of science - Physics department

Abstract:

In this research, the effect of γ -rays from (Cs^{137}) source a time period (21) days on optical properties such as energy gap (E_g) , the optical Transmittance (T%) spectra , optical Absorption spectra (A), optical reflectance spectra (R %), absorption coefficient (α) ,Urbach energy (E_u), extinction coefficient (k) , refractive index(n) and dielectric constants for the lead selenide (PbSe) thin films prepared by the thermal evaporation on a glass substrate   in 5×10^{-5} mbar pressure at room temperature of   thickness (7000 Å) and rate of deposition (1.46 nm/s) . it were seen that all the parameters under investigation affected by gamma irradiation.

تأثير أشعة كاما على الخصائص البصرية لأغشية PbSe الرقيقة المحضر

بطريقة التبخير الحراري في الفراغ

جاسم محمد عبد اللطيف

جامعة ديالى ، كلية العلوم، قسم الفيزياء

الملخص:

تم في هذا البحث دراسة تأثير التشعيع بأشعة كاما لمصدر(Cs^{137}) لمدة (21) يوم على الخواص البصرية المتمثلة بفجوة الطاقة البصرية(E_g)   ، طيف النفاذية البصرية ، طيف الامتصاص البصري (A) ، طيف الانعكاسية البصري (R %) ، معامل الامتصاص(α) ، طاقة اورباخ(E_u) ، معامل الخمود (k) ، معامل الانكسار(n) وثابت العزل بجزأيه الحقيقي والخيالي لأغشية سلايند الرصاص(PbSe) المحضرة بطريقة التبخير الحراري على قواعد زجاجية في ضغط (5×10^{-5} mbar) عند درجة حرارة الغرفة بسمك(7000Å) ومعدل ترسيب (1.46 nm/s) . ولقد وجد أن جميع المعلمات التي تمت دراستها قد تأثرت بأشعة كاما.

**Effect of irradiation on the optical properties of PbSe
thin film prepared by thermal evaporation method
Jasim Mohammed Abid Al-Leteef**

1. Introduction

It is now a well known fact that the exposure of any solid material to ionizing radiations (such as X-rays, gamma rays, beta particles, alpha particles, fission fragments, etc) produces changes in the microstructural properties of the material, which in turn affects the optical and electrical properties.[1]

Studies on the changes in optical properties of thin films irradiated with ionizing radiations yield valuable information's regarding the electronic processes in these materials. High-energy radiations, such as γ-rays, change the physical properties of the materials they penetrate. The changes are strongly dependent on the internal structure of the absorbed substances. It is believed that ionizing radiation causes structural defects (called colour centres or oxygen vacancies in oxides) leading to their density change on the exposure to γ-rays [2].

The presence of such colour centres in a thick film matrix gives rise to changes in both the optical and electrical properties of the material, which can be utilised to assess the radiation dose absorbed [3].

The influence of radiation depends on both the dose and the parameters of the films including their thickness: the degradation is more severe for the higher dose and the thinner films [4].

Lead chalcogenides (PbSe, PbTe, and PbS) are IV-VI narrow band-gap semiconductor over several decade have been motivated by their importance for applications such as IR detectors, photographic plates, photo resisters ,lasers, light-emitting device, photovoltaic's, and high temperature thermoelectric [5-6]. Among the group compounds, Lead selenide (PbSe) thin film is used as a target material in infrared sensor, grating, lenses and various optoelectronic devices [7]. It has a cubic crystal structure (FCC) type ,a lead-grey ,somewhat bluish substance and a direct band gap of 0.27 eV in room temperature ,the refraction index at (3 μm) is (4.54) and the dielectric constant is (280). [8-10] Thus, the PbSe thin films attract attention of many researchers because they are cheap, abundant and they posses semiconducting properties. In the past, several techniques such as thermal evaporation electrodeposition , chemical bath deposition, , photochemical , sputtering, spray pyrolysis, sonochemistry , sonoelectrochemistry , microwave heating, molecular beam epitaxy , electrochemical atomic layer epitaxy and pulsed laser deposition method have been used in the deposition of PbSe thin films.[11,12]

Among the thermal evaporation method is one of the oldest and still most widely used techniques for depositing thin films because of it is cheap, non poisonous, easy method to produce films of extreme purity and , a certain extent of pre-selected structure . [13,14]

The aim of this experimental work is to investigate the changes in the optical properties of PbSe thin film structures under the influence of γ-radiation.

DIYALA JOURNAL FOR PURE SCIENCES

Effect of irradiation on the optical properties of PbSe
thin film prepared by thermal evaporation method
Jasim Mohammed Abid Al-Leteef

2. Experimental

The deposition process of PbSe thin films were done on commercial glass slides with 1mm thick and 2.5×7.5 cm^2 size. Before the deposition they were washed with detergent and a piece of gauze then rinsed many times in distilled water then cleaned by detergent solution then washed by distilled water and finally they were cleaned by using ethanol solution then The ultrasonic was used to clean glass substrates using isopropyl alcohol and finally the substrates were dried by blowing air. Thin films PbSe with weight percentage (0.5) of Pb and Se were grown with a thickness of 0.7 µm on glass substrates at a substrate temperature (T_s) equal to RT by thermal evaporation method using the Edward E306A coating system. The molybdenum boat which was used for film deposition with a high purity (99.999%) and covered with baffle. since the vacuum pressure during evaporation was about 2×10^{-5} torr and the deposition rate is about 1.46 nm/s.

The distance between the boat and substrate was kept at 15 cm. The substrate temperature was measured and controlled using digital thermometer.

A disc-type Cs^{137} gamma radiation source was used to expose PbSe thin films under Investigation for seven days to dose of about 18 Gy at room temperature.

The optical transmission and absorption spectra for unirradiated and irradiated PbSe films deposited at room temperature were obtained in infrared region with 2.5 µm to 5 µm using PYE UNICAM(Philips) , UV- spectrophotometer (Model: PU 9712). The measurements were done in the wavelength scanning mode under the infrared radiation(IR).

3. Results and discussion

The optical properties of a semiconductor can be defined as any property that involves the interaction between electromagnetic radiation or light and the semiconductor, including absorption, diffraction, polarization, reflection, and scattering effects. The best values of the band gap are obtained by the measurement of optical absorption[15]. Knowledge of optical constant of a film from a given material is basic importance in determining the characteristics of light transmission of the film. Such knowledge of the optical parameters of the material is of importance when designing devices through which electromagnetic radiation absorbed or transmitted[16].Therefore, the optical Transmittance (T%) and optical Absorption (A) spectra of unirradiated and irradiated PbSe films deposited at R.T in the spectral range(2.5-5 µm) are shown in Figure (1).

**Effect of irradiation on the optical properties of PbSe
thin film prepared by thermal evaporation method
Jasim Mohammed Abid Al-Leteef**

The percentage of transmittance peak for unirradiated and irradiated PbSe films deposited at R.T are decreases from 32% to 25% in infrared region respectively . We notice also that the transmittance (for both curves) decreases with uniform on the exposure to γ -rays and shifted toward longer wavelengths. This behavior could be explained as follows ; an increasing roughness of the irradiated thin films contributed to the drastic decrease of optical transmittance[17].
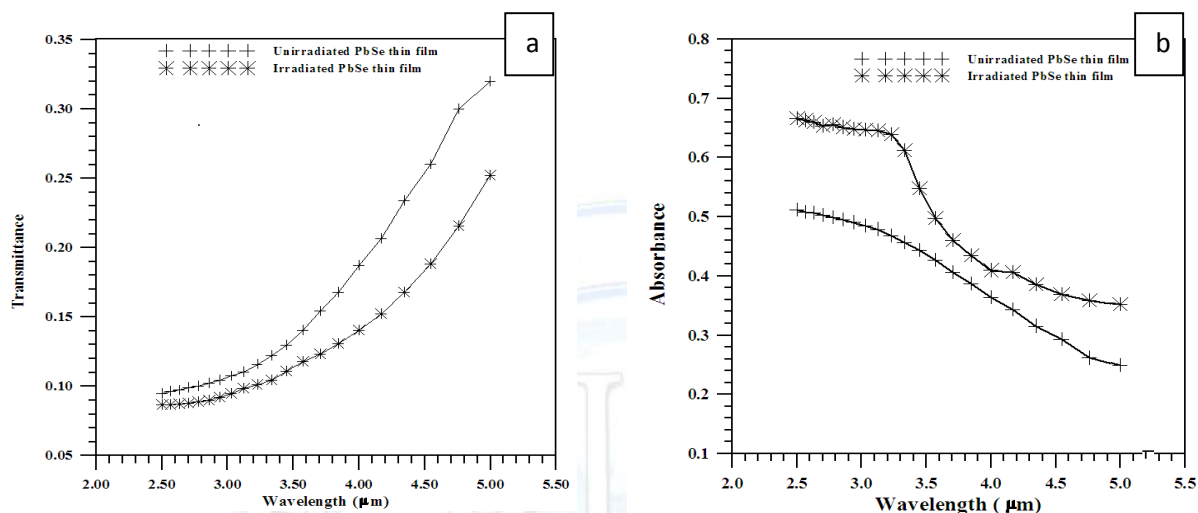
Figure(1) (a) Transmittance[%] versus Wavelength. (b) Absorbance versus Wavelength

It is observed that the Absorbance e (for both curves) decreases at the spectral region of fundamental absorption as a function of wavelength (μm) from (2.5 to 5μm).

The spectral behavior of these films shows that the absorption edge shift to shorter wavelength (higher energy) for the irradiated thin films. In this region the incoming photons have sufficient energy to excite electrons from the valence band to the conduction band and thus these photons are absorbed within the material to decrease the transmittance.[18]

Fig. 1c is a plot of reflectance as a function of wavelength for PbSe thin films in this work. In general the film shows the reflectance is below 43.3% and decreases with irradiation. This is occurs as a result to transmittance decreasing according to the relation[19]:

R=1- T-A …………………………………………………..… (1)

DIYALA JOURNAL FOR PURE SCIENCES

**Effect of irradiation on the optical properties of PbSe
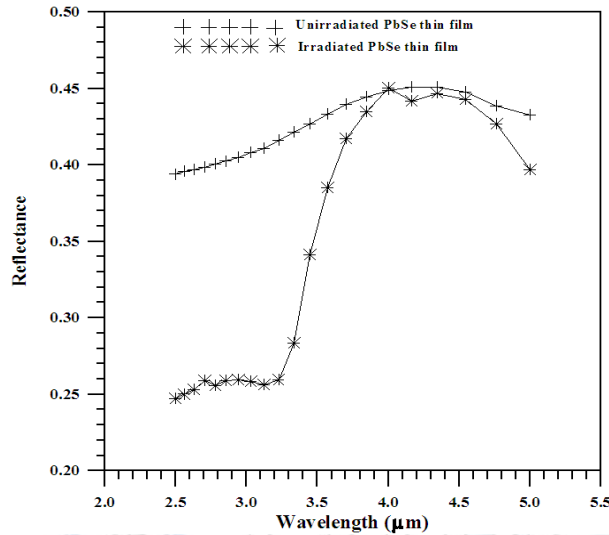thin film prepared by thermal evaporation method
Jasim Mohammed Abid Al-Leteef**

Fig. 1c. Reflectance vs. wavelength for PbSe

The variation of absorption coefficient(α) with wavelength is shown in the Fig.(2). Both films show higher absorption(or shift) on the shorter wavelength(or higher energy) side, this is attributed to increase the defect states which leads to increase absorption coefficient.
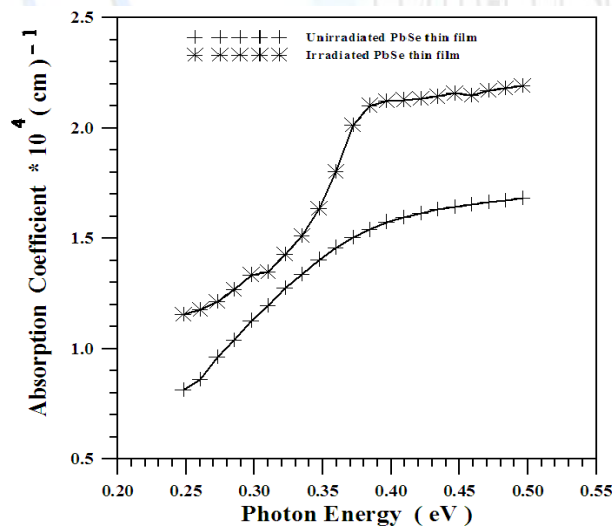

Figure (2) Absorption Coefficient as a function of Photon Energy (eV).

Clearly, exposure to gamma radiation produces considerable change in the shape of the optical absorption spectrum around the absorption edge. From an optical absorption curve, the

DIYALA JOURNAL FOR PURE SCIENCES

**Effect of irradiation on the optical properties of PbSe
thin film prepared by thermal evaporation method
Jasim Mohammed Abid Al-Leteef**

values of the absorption coefficient, α_v, for different values of the photon frequency, were calculated using the relationship[20]:

$$\alpha_v = 2.303 \, A_v / t \quad \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots \quad (2)$$

where A_v denotes the absorbance corresponding to the photon frequency(v) and (t) the thickness of the thin film. The absorption coefficient near the band edge shows an exponential dependence on the photon energy and obeys the Urbach's empirical formula [21] :

$$\alpha_v = \alpha_o \, e^{hv/\Delta E} \quad \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots \quad (3)$$

where α_o is a constant, h the Planck's constant and ΔE the energy width of the band tails of localized states. The graphs of $\ln\alpha$ vs photon energy(hv) were plotted for unirradiated and irradiated PbSe films deposited at R.T and the values of ΔE were calculated from the reciprocal of the slopes of the straight-line portions of these plots as shown in figure 3. The energy width of the band tails of localized states (ΔE) has been found to increase with the gamma radiation from(0.137 eV to 0.183 eV) for unirradiated and irradiated films respectively . This increase in the energy width of the band tails of localized states can be attributed to the induced defects due to the exposure to gamma radiation.
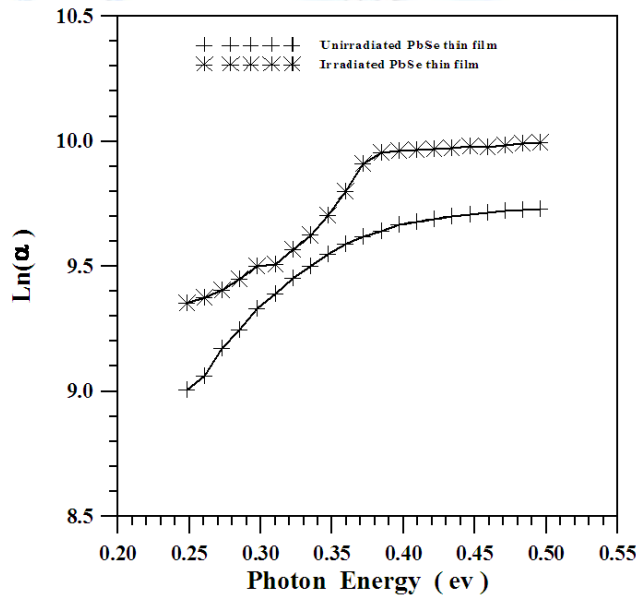

Figure (3) Ln(α) as a function of Photon Energy (eV).

The optical band gap was calculated using the Tauc [22] relation given by the formula

$$\alpha hv = A(hv - Eg)^n \quad \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots \quad (4)$$

DIYALA JOURNAL FOR PURE SCIENCES

**Effect of irradiation on the optical properties of PbSe
thin film prepared by thermal evaporation method
Jasim Mohammed Abid Al-Leteef**

Here hν is the photon energy, α is the absorption coefficient , Eg is the optical band gap, A is a constant and n =1/2 for direct band gap material. We plotted $(\alpha h\nu)^2$ vs. photon energy in the wavelength range 2.5–5 μm , the values of the optical band gap are determined by extrapolating the linear part of $(\alpha h\nu)^2$ vs. energy axis for direct allowed transitions, as shown in figure(4).

.
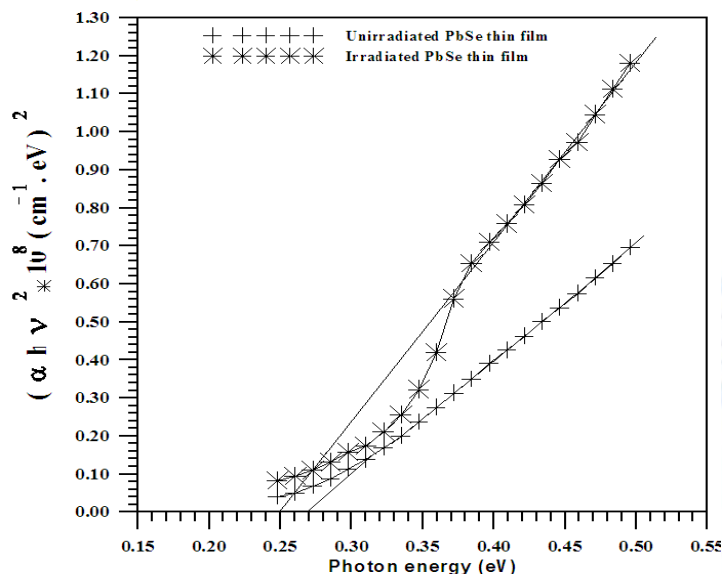
Figure(4) Band gap E_g estimation for PbSe films.

The optical band gap was shifted from 0.27 eV to 0.25 eV due to irradiation . Gamma doses cause the breaking of bonds, leading in turn to the increase of dangling bonds and of defects, as well as the trapping of the generated carriers. This may be the cause for the increase in band tail width(Δ E), and then decrease energy gap.
Refractive index is one of the fundamental properties for an optical material, because it is closely related to the electronic polarizability of ions and the local field inside materials. The evaluation of refractive index of optical material is important for many applications especially in optical devices .It is clear from figure (5) for the irradiated film ,that there are two regions of refractive index (n) spectrum. The first region occurs at (λ=2.5-4 μm) and the second region starts where λ exceeds(4 μm).The refractive index (n) increases in the first region ,results from the electronic transitions of valence band and the impurities(or increase of the compactness of the films) and the transitions of energy bands. These values are reduced sharply with increasing in wavelengths. In the second region of spectrum, the variation of refractive index (n) with wavelengths becomes slight decreases and its values are very close at cut-off wavelength. It can be calculated from the following equation [23,24]:

$$ n = \left[(\frac{1 + R}{1 - R})^2 + (k^2 + 1) \right]^{1/2} + \frac{1 + R}{1 - R} \dots\dots\dots\dots\dots\dots\dots\dots\dots\dots\dots(5) $$

**Effect of irradiation on the optical properties of PbSe
thin film prepared by thermal evaporation method**
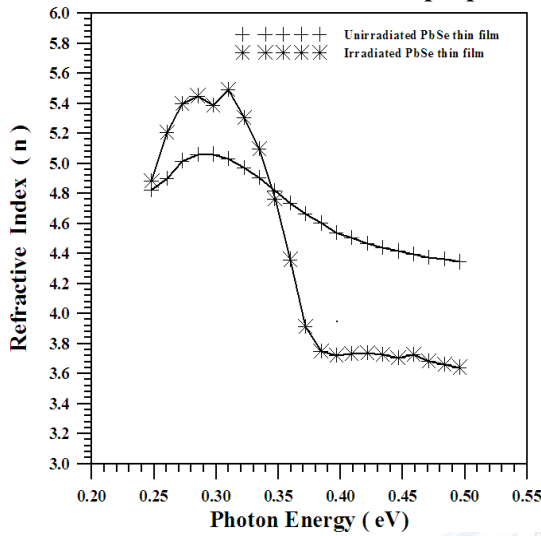
**Figure(5) Refractive Index vs. Photon
energy.**
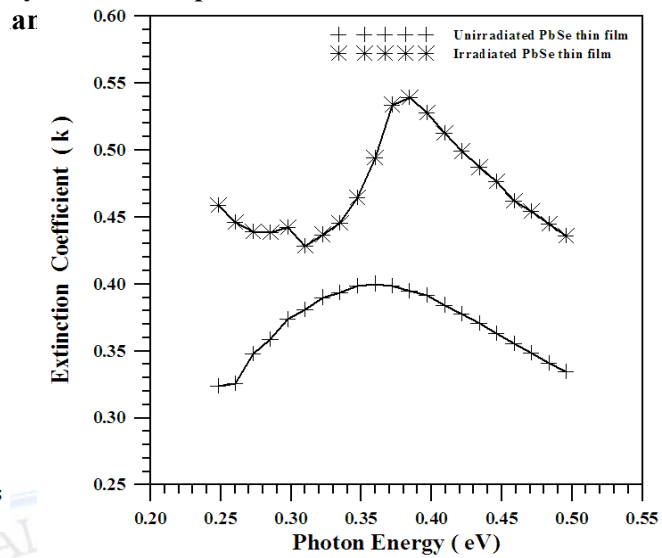
**Figure(6) Extinction coefficient vs.
Photon energy.**

The extinction coefficient (k) represents the imaginary part of the refractive index , which is related to the exponential decay of the wave as it passes through the medium can be determined by using equation [25,26]

$$ k = \frac{\alpha\lambda}{4\pi} $$ ……………………………….. ……………………………………(6)

λ: is the wavelength of the incident photon.

The figure (6) shows the extinction coefficient (k) as a function of photon energy, the (k) for the unirradiated PbSe films less than (or decreases) the irradiated PbSe films ;This is attributed to the same reason mentioned previously in the absorption coefficient because the behavior of k is similar to α.
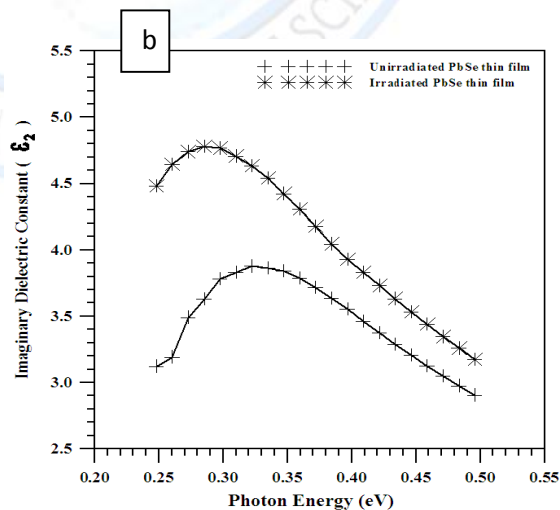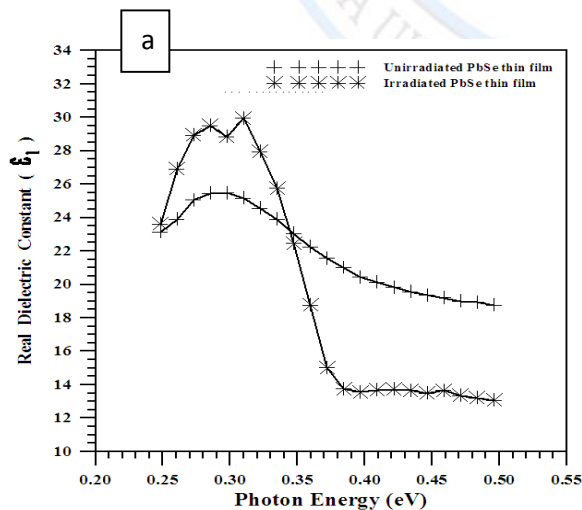

Figure (7-a&b)The (ε_1) and **The (ε_2) as a function of Photon Energy (eV).**

DIYALA JOURNAL FOR PURE SCIENCES

**Effect of irradiation on the optical properties of PbSe
thin film prepared by thermal evaporation method
Jasim Mohammed Abid Al-Leteef**

The dielectric constants consists of real part (ε_1) and imaginary part (ε_2), the variations of them with photon energy as shown in figure (7-a&b) and can be calculated by using equations[27]:

$$\varepsilon_1 = n^2 - k^2 \quad \text{……………………………………………………...(7)}$$

$$\varepsilon_2 = 2nk \quad \text{………………………………………………………….(8)}$$

The variation of ε_1 and ε_2 with the increase of the wavelength of the incident radiation is due to the change of reflectance and absorbance [28]. The behavior of ε_1 is similar to that of the refractive index because of the smaller value of k^2 compared with n^2, while ε_2 mainly depends on the k value, which are related to the variation of absorption coefficient. ε_2 represents the absorption of radiation by free carriers[29]. It is observed that ε_1 increases and decreases with irradiation, and this attributed to the same reason mentioned previously for the refractive index, while ε_2 increases with increasing the irradiation and this is due to the similar interpretation discussed previously for the extinction coefficient.

Conclusions

We can arrive at the following conclusions after the irradiation by gamma ray on PbSe thin film:

- ❖ Decreasing the transmittance spectral and the reflectance spectral caused by increasing the defect states which leads to the two spectral curve shifted toward the longer wavelengths,
- ❖ Increasing the Absorbance caused by increasing the defect states which leads to the position of the fundamental absorption edge shifted toward the shorter wavelength (higher energy),
- ❖ Increasing the absorption coefficient and the extinction coefficient,
- ❖ Increasing the energy width of the band tails of localized states (ΔE) this is attributed to increase the defect states which leads to decreasing the energy gap from 0.27 eV to 0.25 eV ,
- ❖ In general , decreasing the dielectric constants consists of real part (ε_1) and imaginary part (ε_2).

References

[1] A. M. Ibrahim and L. I. Soliman,"Rad. Phys. & Chem.", 53 ,469 ,(1998).

[2] R.Y.Zhu, 413, 297-311,(1998).

[3] A. Arshak, S. Zleetni and K. Arshak," Sensors",2, (2002)

[4] E. Tanassova, A. Paskaleva , R. Konakova , D. Spassov, V. F. Mitin ,32,553-562, (2001).

[5] Khang Hoang ,S.D.Mahanti, and Puru Jena,"Phys.Rev.",B76,115432,(2007).

DIYALA JOURNAL FOR PURE SCIENCES

**Effect of irradiation on the optical properties of PbSe
thin film prepared by thermal evaporation method
Jasim Mohammed Abid Al-Leteef**

[6]   S.Prabahar,N.Suryanarayanan,K.Rajasekar,S.Srikanth,"Chalcogenide Letter", Vol.6,No.5,P.203-211.(2009).

[7]   M.S. Ali, K.A. Khan, M.S.R. Khan, Phys. Status. Solidi. (a) 149,611, (1995).

[8] R. A. Smith , " Semiconductors " , 2nd edition , Cambridge University Press , (1987)

[9]   D.M. Chizhikov and V.P. Shchastlivyl ," Selenium and Selenides",Collets(1968).

[10] D .R. Lide , ((Handbook of Chemistry and Physics)) , LRC Press , Boca Raton ,
     Florida USA ,77th edition , (1996-1997).

[11] A. Kassim., .T .W. Tee, D.K.Abdullah, M. J. Haron, H. S.Min, S., Monohorn   and
N. Saravanan ," J.Kimia",Vol.4,No.1,P.1-6,(2010).

[12] K.W. Li, X.T. Meng, X. Liang, H. wang, H. Yan,"J. Sol. Stat. Elect.", Vol.10,
     P.48- 53,(2006).

[13] L. Eckertova , ((Physics of Thin Films)) , Plenum presses, New York and
London (1977).

[14] R. W. Berry , P. M. Hall & M. T. Harris , ((Thin Film Technology)) , New York   ,
Cincinnati , Toronto , London , Melbourne , (1979).

[15] C. Kittel, "Introduction to Solid State Physics", 8th ed., John Wiley and Sons, Inc., New
     York, (2005).

[16] R. J. Elliot and A. I. Gibson, "An Introduction to Solid State Physics and Application",
     1st ed., Macillian Inc. (1974).

[17] N. Tigau , V. Ciupina , G. Prodan , G. I. Rusu , C. Gheorghies , E. Vasile , J.
Optoelectron. Adv. Mater., 5, 907 ,(2003).

[18] A.N. Banerjee1, C.K. Ghosh & S. Das, K.K. Chattopadhyay .Physica B 370 ,
264–276. (2005).

[19] G. Hass , ((Physics of Thin Films)) , Volume 1, Academic Press, 1963.

[20] T.K.Malty & S.I.Sharam ,"Bull. Mater . Sci.",Vol.31 ,No.6 ,pp.841-846 ,(2008).

[21] F. Urbach , " Phys. Rev." ,92 ,1324,(1953).

[22] J Tauc, "Amorphous and liquid semiconductrors" edited by J Tauc (Plenum
Press, New York, p. 159,(1974).

[23] J. C. Mainfacier, J. Gasiot and J. P. Fillard, "J. Phys. E. Sci. Instrum.", 9, (1989).

[24] K. L. Chopra, "Thin Film Phenomena", McGraw-Hill, Inc., New York, (1969).

[25] J. I. Pankove, "Optical Processes in Semicoductors", Prentice – Hall, Inc.,
     Englewoodcliffs, New Jersy (1971).

[26] R. A. Grenier "Semiconductor Device", Electronic Energy Series, McGraw-Hill, Book
     Co. Inc. (1961).

[27] A. H. Clark, "Polycrystalline and Amorphous Thin Films and Devices", Academic Press,
     New York, (1980).

[28] G.I. Rusu, P. Prepelita and N. Apetraoei, "Journal of Optoelectronics and Advanced
     Materials", Vol.7, No.2, P. 829, (2005).

[29] P. Burrows and S. Forrest, "J. Appl. Phys.", 79, 10, 7991, (1996).