



Enhancement of the Dielectric and Nonlinear Optical Properties of PbSe Nanomaterial Thin Films with Different Contents of Polyethylene Glycol



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Samia A. Gad,^a Ahmed Abdel Moez,^b Gehan M. Mahmoud,^b Fatma S. Terra,^{b,*}

^aSemiconductor Laboratory, Solid State Physics Department, Physics Research Division, National Research Centre, 33 El-Bohouth Street, Dokki, Giza 12622, Egypt.

^bSolid State Electronics Laboratory, Solid State Physics Department, Physics Research Division, National Research Centre, 33 El-Bohouth Street, Dokki, Giza 12622, Egypt.

Abstract

PbSe nanocrystals doped with poly ethyleneglycol (PEG) were prepared using hydrothermal method. The reflectance spectra (R) were measured for these samples, and also refractive index (n) values were calculated, which increase with PEG ratio. Both of (E_d) and (E_o) increase with PEG content, while (N/m^*) ratio decreased with PEG content. While all of (M_{-1}), (M_{-3}) and static refractive index (n_o), were determined. (ϵ^{\parallel}) and (ϵ^{\perp}) peak position decrease with photon energy ($h\nu$). Also both of (σ_1) and (σ_2) decrease with ($h\nu$), while ($\chi^{(1)}$) increases with ($h\nu$) for all samples. The nonlinear optical parameters such as, (n_2) and ($\chi^{(3)}$) decrease with PEG, as a result of increasing the sample density with PEG. (β_c), were determined theoretically. Both electrical susceptibility ($\chi_{(e)}$) and relative permittivity ($\epsilon_{(r)}$) has blue shift with ($h\nu$) for all samples. (N_c/m^*_e (cm^{-3})), (N_v/m^*_h (cm^{-3})) were calculated theoretically.

Keywords: PbSe nanocrystals with different PEG contents; optical conductivity; dielectric properties; nonlinear optical properties; semiconducting properties.

1. Introduction

In the recent time the most common precision and advanced industries depend mainly on nanomaterials [1] as a result of their wide electronic and optoelectronics applications, as a result of their sorption[2-5], catalytic[6-8], optical[9], electrical, and other special properties [10-11]. Lead chalcogenides are promising materials due to their narrow band gap [12]. PbSe has narrow energy band gap (0.27 eV) [13], which increases to (~1.5 eV) by decreasing crystallite size [14]. PbSe nanocrystals have a large electronic applications such as, field effect transistors [15],

infrared detectors [16], thermoelectric material [17]. PbSe thin films had been prepared with different methods such as, electrodeposition [18], Inert gas condensation [19], electron beam evaporation [20], Chemical bath deposition [21]. PbSe thin films had a polycrystalline structure [22-24] with cubic structure and lattice constant ($a = 6.124 \text{ \AA}$) [25]. The physical and optical properties of PbSe thin films were studied by many authors [26-35], PbSe thin films had band gap of 1.18 eV[26], with range (1.5-1.8 eV)[27], (1.5 and 1.9 eV)[28] energy gap decreased with increasing film thicknesses [29-31], it decreased from (1.89-1.60 eV) for films with thickness [31]. The electrical properties of PbSe thin films were studied [22, 36-39]. The electrical resistivity decreased with film thickness

*Corresponding author e-mail: aam692003@yahoo.com; (Ahmed Abdel Moez).

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[22], and applied pressure [38], Ac conductivity is thickness independent [39]. The dopant effect on physical properties for PbSe thin films were investigated [40-42]. It was found that, PbSe thin films have stable photo response to infrared light with Te dopant [41], electrical resistivity decreases with increasing Te ratio [42]. PbSe thin films prepared by different contents of PEG were studied [43]. It was found that, both of energy gap and electrical resistivity increase with PEG content. In this paper we investigated the influence of PEG contents on nonlinear optical properties of PbSe nanomaterial thin films, such as nonlinear refractive index, nonlinear absorption coefficient, third order nonlinear optical susceptibility, and finally the semiconducting results.

2. Experimental work

PbSe nanocrystals were prepared by a simple hydrothermal method. In a typical synthesis, different contents of poly- ethylene glycol (20, 60, 80 and 100 mg) and 4.8 g of sodium hydroxide were added to 100 mL of de-ionized water. After few minutes of stirring, 2 mM of lead acetate trihydrate were added to the solution and stirred until the reactants were dissolved completely. 20 mL of hydrazine hydrate (20.000) was finally added to the solution and transferred into a Teflon-lined autoclave. The sealed vessel was kept in a furnace at a temperature of 100C for 12 h and then cooled down to room temperature. The product was washed several times with double distilled water. Thin films have been prepared by thermal evaporation technique using a high vacuum coating unit (Edwards E306A) under vacuum of about 5×10^{-4} Pa. The films were deposited onto well-cleaned optically flat quartz substrates for the optical measurements and onto well-cleaned glass substrates for the structural and electrical measurements. During deposition, the substrates were kept at room temperature. The film thickness (≈ 120 nm) and deposition rate (1.5–2 nm/s) were measured during the evaporation process by using a quartz crystal thickness monitor. The transmittance and reflectance spectra of the films were recorded by a double beam spectrophotometer JASCO model (V-570 UV-Vis-NIR). The measurements were performed in the wavelength range 500–2500 nm.

3. Results and Discussions

3.1. Optical results

The measured optical transmittance (R) for these samples is shown in figure 1a. (n) for these films were calculated using the following equation [44]

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

Figure 1b. shows (n) dependence on the (λ) for PbSe films with different content of PEG, the behavior of n with λ is the same for all samples, but the n values increase with PEG content, due to increasing the sample density [45].

3.2. Dielectric, optical conductivity and linear optical susceptibility results

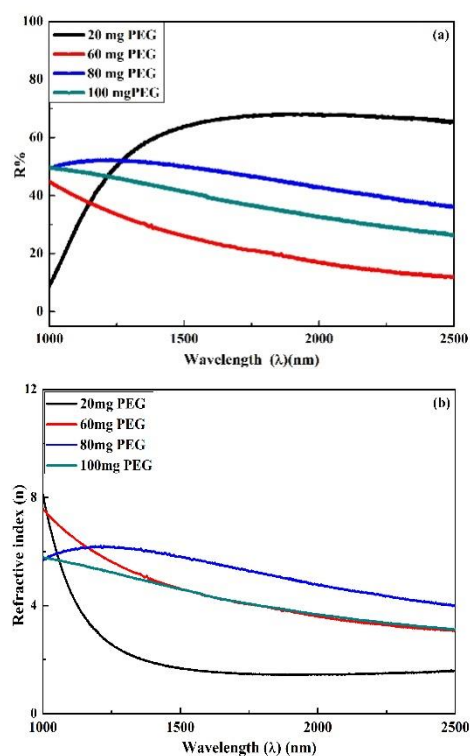


Fig. 1. Relation between wavelength (λ) and (a) Reflectance, (b) dependence of refractive index (n) for PbSe films Prepared with different content of PEG.

The single oscillator for these sample can be as follow [46]:

$$n^2(E) - 1 = \frac{E_o \cdot E_d}{E_o^2 - E^2} \quad (2)$$

Where E is the photon energy (hv), E_o is the oscillator energy and E_d is the dispersion energy.

Both of E_o and E_d increase with PEG content. figure 2a shows the relation between $(n^2-1)^{-1}$ vs. $(hv)^2$. $(n^2-1)^{-1}$ increases as the PEG. While, figure 2b. shows, the relation of n^2 and λ^2 to determine (n/m^*) and the lattice dielectric constant (ϵ_L) as follow [47]:

$$n^2 - k^2 = \epsilon_L - \left(\frac{eN}{4\pi c^2 \epsilon_o m^*} \right) \lambda^2 \quad (3)$$

Where ϵ_o is the permittivity of free space, e is the charge of electron, k is absorption index of these films respectively, which was determined in a previous work [43], and c is the speed of light, so the values of (N/m^*) and ϵ_L are shown in table 1. From this table it was noticed that (N/m^*) decreases with PEG, due to increasing the sample density. The first order of moment (M_{-1}) and the third order of moment (M_{-3}) are that central and standardized motion of electrons respectively and were derived from the relations [47]:

$$E_o^2 = \frac{M_{-1}}{M_{-3}} \quad (4)$$

$$E_d^2 = \frac{M_{-1}^3}{M_{-3}} \quad (5)$$

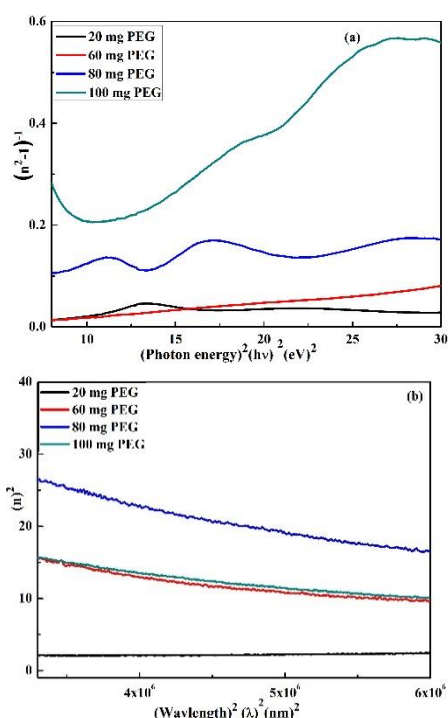


Fig. 2. (a) The relation of $(n^2-1)^{-1}$ and $(hv)^2$, (b) relation between (n^2) and (λ^2) for PbSe thin films with different contents of PEG.

The oscillator strength (f) was calculated as follows [48]:

$$(6)$$

$$f = E_o \cdot E_d$$

The static refractive index n_o , which is the medium ability to refract the light depending on the electron oscillations, and was determined as [49]:

$$n_o = \left[\left(\frac{E_d}{E_o} \right) + 1 \right]^{0.5} \quad (7)$$

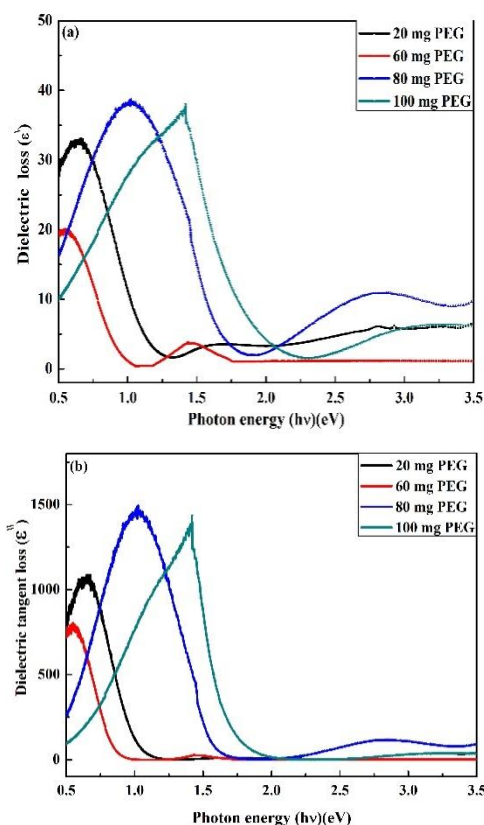


Fig. 3. (a) Dependence of dielectric loss (ϵ') on (hv) , (b) dependence of dielectric tangent (ϵ'') on (hv) for PbSe thin films with different contents of PEG.

The values of (n_o) increase with PEG content as shown in table 1, because of increasing the samples density. Both of (ϵ') and (ϵ'') for these films were determined as [50]:

$$\epsilon' = (n^2 + k^2) \quad (8)$$

$$\epsilon'' = \left[(n^2 + k^2)^2 - (n^2 - k^2)^{0.5} \right] \quad (9)$$

Dependence of Both of (ϵ') and (ϵ'') on (hv) is shown in Figs 3(a,b), peak position for (ϵ') and (ϵ'') increases with PEG, due to the decreased of electron motilities. The optical conductivity was calculated from the following equations [51]:

$$\sigma_1 = \left(\frac{\epsilon'' \cdot c}{2\lambda} \right) \quad (10)$$

$$\sigma_2 = \frac{(1 - \epsilon') \cdot c}{4\lambda} \quad (11)$$

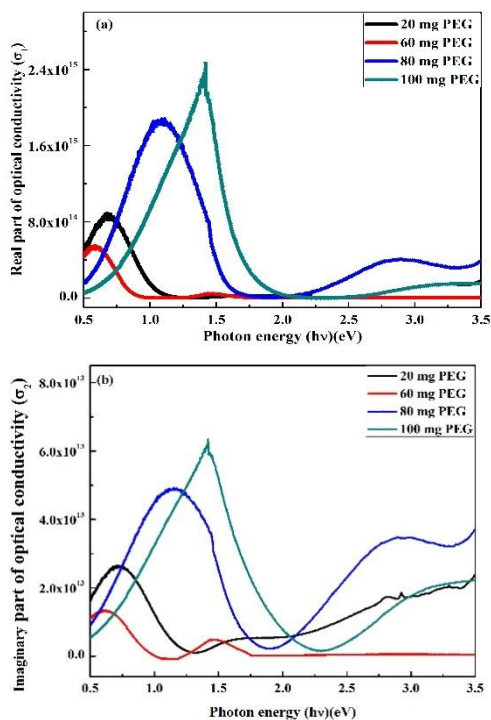


Fig. 4. (a) Influence of $(h\nu)$ on real part of optical conductivity (σ_1), (b) influence of $(h\nu)$ on imaginary part of optical conductivity (σ_2) for PbSe thin films with different contents of PEG.

figure 4(a and b) show, the both of (σ_1) and (σ_2) dependence on $(h\nu)$ for these films. ($\chi^{(1)}$) describes the response of the material to an optical wave length, ($\chi^{(1)}$) was determined as [52]:

$$\chi^{(1)} = \frac{(n^2 - 1)}{4\pi} \quad (12)$$

The dependence of ($\chi^{(1)}$) on $(h\nu)$ is shown in Fig.5a, ($\chi^{(1)}$) increases with PEG, this means that, the response for optical excitation increase with PEG.

Nonlinear optical properties

The non-linear refractive index (n_2), is change in the refractive index in proportion to the optical intensity [53], n_2 was determined as [54-55]:

$$n_2 = \frac{(12\pi\chi^{(3)})}{n_o} \quad (13)$$

The dependence of n_2 on (λ) is shown in figure 5b. (n_2) decrease with (λ) , and also decrease with PEG, as a result of increasing the sample density.

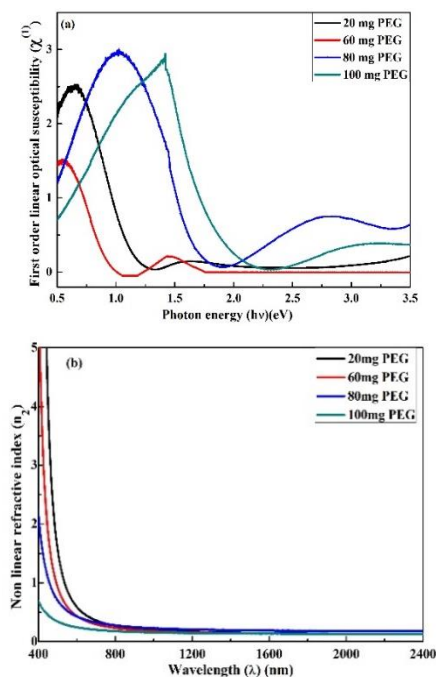
An important parameter to assess the degree of nonlinearities is ($\chi^{(3)}$), which was as [56]:

$$\chi^{(3)} = A \left[\frac{E_o \cdot E_d}{4\pi(E_o^2 - (h\nu)^2)} \right]^4 \quad (14)$$

Where $A = 1.7 \times 10^{-10}$ e.s.u [57]. The effect of $(h\nu)$ on $\chi^{(3)}$ is shown in Fig.5c. It was noticed that, ($\chi^{(3)}$) increases with $(h\nu)$, this is due to the fact that, the light deflection increase with $(h\nu)$.

On the other hand, the nonlinear absorption coefficient (β_c) is determined as follows [57]:

$$\beta_c = \frac{48 \cdot \pi^3 \cdot \chi^{(3)}}{n^2 \cdot c \cdot \lambda} \quad (15)$$



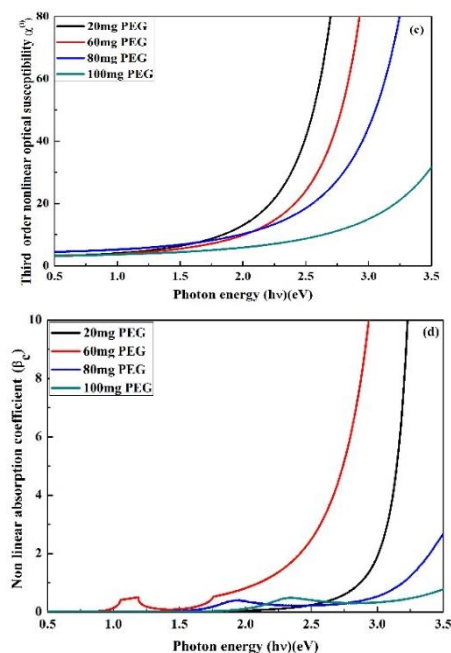


Fig. 5. (a) Relation between first order of linear optical susceptibility ($\chi^{(1)}$) and $(h\nu)$, (b) Relation between nonlinear refractive index (n_2) and wavelength (λ), (c) Dependence of third order of nonlinear optical susceptibility ($\chi^{(3)}$) on $(h\nu)$, (d) The influence of $(h\nu)$ on nonlinear absorption coefficient (β_c) for PbSe thin films with different contents of PEG.

Figure 5d. shows the influence of $h\nu$ on (β_c) . It is observed that, (β_c) increases with increasing $h\nu$. Because of large number of excited electrons overcome the band gap with $(h\nu)$

3.3. Electrical results

Electrical susceptibility ($\chi_{(e)}$) was determined using the following relation [58]:

$$\chi_{(e)} = \frac{(n^2 - k^2 - \epsilon_o)}{4\pi} \tag{16}$$

Figure 6a. shows the relation between ($\chi_{(e)}$) and $(h\nu)$ of these investigated samples. ($\chi_{(e)}$) increase with $(h\nu)$. This is due to; the electron mobility increases with $(h\nu)$. The relative permittivity (ϵ_r) was calculated using the following relation [59]

$$\epsilon_r = (\chi_e + 1) \tag{17}$$

The dependence of (ϵ_r) on (λ) is shown in figure 6b. (ϵ_r) increase with $(h\nu)$ for all these samples; this could be attributed to, the electron mobility increases with $(h\nu)$.

Table 1. The determined values for both N_v , N_c

3.4. Electronic results

The density of states (DOS) was calculated as follows [60]: -

$$N_v = 2 \left[\frac{(2\pi m^*_h KT)}{h^2} \right]^{3/2} \tag{18}$$

$$N_c = 2 \left[\frac{(2\pi m^*_e KT)}{h^2} \right]^{3/2} \tag{19}$$

The determined values for both N_v , N_c were shown in table 1.

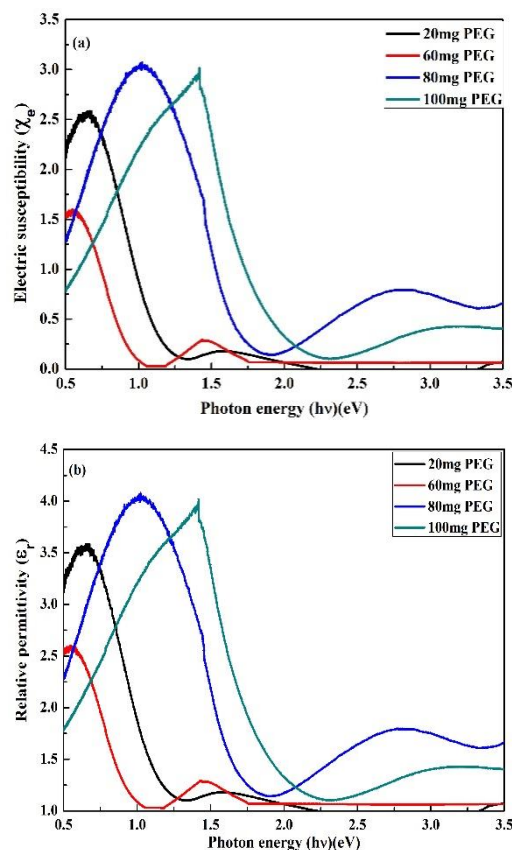


Fig 6. The influence of $(h\nu)$ on both of (a) electrical susceptibility (χ_e), (b) relative permittivity (ϵ_r) for PbSe thin films with different contents of PEG

Table 1. The determined values of PbSe thin films with different contents of (PEG), such as, ϵ_L , E_o , E_d , M_{L-1} , (f) , (n_o) , (N/m^*) , (N_C/m^*_h) and (N_v/m^*_e) .

| PbSe thin films with PEG content (mg) | lattice dielectric constant ϵ_L | Oscillation energy E_o (eV) | Dispersion energy E_d (eV) | M_{-1} (eV) | M_{-3} (eV) | Field strength (f) (eV) ² | n_o | N/m^* | N_C/m^*_h | N_V/m^*_e |
|---------------------------------------|--|-------------------------------|------------------------------|---------------|---------------|--|-------|---------|-------------|-------------|
| 20 | 2.00 | 3.60 | 5.20 | 4.33 | 2.28 | 18.72 | 1.56 | 1.1E+51 | 9.3E+20 | 4.10E+21 |
| 60 | 15.00 | 3.90 | 5.60 | 4.67 | 2.37 | 21.84 | 1.56 | 8.2E+50 | 7.3E+20 | 3.50E+21 |
| 80 | 25.00 | 4.50 | 7.10 | 5.65 | 2.66 | 31.95 | 1.60 | 7.2E+50 | 8.6E+20 | 3.30E+21 |
| 100 | 15.00 | 5.30 | 7.80 | 6.43 | 2.79 | 41.34 | 1.57 | 6.9E+50 | 8.2E+20 | 3.18E+21 |

4. Conclusions

Both of R and n of PbSe thin films increase with different PEG contents (20, 60, 80 and 100 %), as a result of increase the sample density. The determined values of (E_d) and (E_o) were (E_o (3.60-5.30 eV), E_d (5.2 -7.8 eV) increase for PbSe thin films with PEG content, due to increase the electron variation, and the wave dispersion through a medium, and also (N/m^*) decreases with PEG content, as a result of increasing m^* with PEG of these sample. The values of (M_{-1}) and (M_{-3}) also increase with increasing PEG content, due to increase both of E_o and E_d , the medium ability to refract the light increase with PEG ratio, causing increase of n_o . Both of (ϵ^l) and (ϵ^ll) increases with ($h\nu$), and the peak maximum had a blue shift with PEG content, this is due to the decrease of electron motilities. This result is also similar to both of (σ_1) and (σ_2). ($\chi^{(1)}$) increased with ($h\nu$), for these samples, and the peak position had blue shift with PEG, which is meaning that: these films had a high response for changing their properties with PEG ratios, while both of ($\chi^{(3)}$) and (βc) increased with ($h\nu$), this is due to: when ($h\nu$) increased the deflected light increased and also the number of excited electrons which overcome the band gap. The increase of electron mobility leads to increase both of ($\chi_{(e)}$) and (ϵ_r) with ($h\nu$) for all these samples. The determined values for both of (N_v) and (N_c) increase the effective mass of these samples with PEG contents.

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