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EMISSION CROSS SECTION AND LUMINESCENCE SPECTROSCOPY OF SAMARIUM OXIDE DOPED TELLURITE GLASSES

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Tellurite glasses with composition $75TeO_2$-$12.5Nb_2O_5$-12.5ZnO- 3000ppm Sm_2O_3 was prepared by using a conventional melt quenching method. The optical properties of this glasses system investigated by using UV-Vis-NIR absorption spectra in the range from 200 to 2500 nm and linear refractive indices *(n)* at different wavelength was estimated. From the absorption edge studies, the value of optical band gap *(E_{opt})* was determined**.** Moreover, the nonlinear refractive index *(n_2)*, third-order nonlinear susceptibility ($\chi^{(3)}$), and nonlinear absorption coefficient, *(β)*, were observed. It is noticed that the nonlinear parameters*n_2, $\chi^{(3)}$ and β* increase by decreasing the value of optical band gap *(E_{opt})***.** The gain cross-section of laser transition level from $^6H_{5/2} \rightarrow {}^6F_{1/2}$was obtained. This glass has the effective emission cross section bandwidth (41 nm) and large stimulated emission cross-section ($1.09x\ 10^{-20}cm^2$). Spectroscopic properties indicate that this glass doped with Sm^{3+} is a promising candidate for optical applications.

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Keywords: Tellurite glasses; Sm^{3+}ions; UV- Vis- NIR, Luminescence analysis; Gain cross section

1. Introduction

Tellurite glasses have many advantages in comparison with other conventional glasses due to their high transparency in the mid infrared region (5 –11μm), high linear and non-linear refractive index**,** low-melting temperature**,** good mechanical stability, chemical durability, lowest cut-off phonon energy and low crystallization rate so these glasses are known as very good hosts for rare earth ions**.** Samarium have many promising characteristics such as it is possesses strong fluorescence intensity, rich energy levels and large emission cross section [1]**.** So the host glass containing Sm^{3+} ions is a promising material for many applications such as solid state lasers, under sea communication, high-density optical storage materials, color displays, temperature sensors and medical diagnostics [2, 3].

From the literature, addition of ZnO and Nb_2O_5 can improve optical nonlinearity, chemical durability, verification and thermal stability of the glasses Nb_2O_5- TeO_2, ZnO-TeO_2, and PbO-Nb_2O_5- TeO_2, TeO_2- Nb_2O_5- ZnO [4-7]

Many systems of glass doped with Sm_2O_3were studied and reveals high values of stimulated emission cross-section, optical gain and gain bandwidth PbO/ TeO_2 / P_2O_5/ ZnO/ $BaCO_3$/ Sm_2O_3**,** TeO_2/ RO/ ZnO/ Nb_2O_5/ B_2O_3/ Sm_2O_3(where R=Mg, Ca and Sr),B_2O_3/ PbO/ PbF_2/ Bi_2O_3/ ZnO/ Sm_2O_3 and TeO_2/ Nb_2O_5 / ZnO/ Er_2O_3 [8- 11]**.**

In this paper a new system of glass TeO_2 / Nb_2O_5 / ZnO / Sm_2O_3 introduced as a novel optical material for the development of lasers and photonic devices

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2. Experimental Work

Glasses with the compositions 75TeO_2-12.5Nb_2O_5-12.5ZnO-3000ppm Sm_2O_3 (denoted by TNZ: Sm) were prepared by the conventional melt quenching technique where specified weights of raw materials (powders) are mixed and given in a covered platinum crucible and heated in a melting furnace to a temperature of 900 °C for 30min; the melt was mechanically stirred after 20 mints and it is being returned to the furnace for 10 mints. The viscous melt was cast in a graphite mold. Subsequently, the sample was transferred to an annealing furnace for 2h at 350 °C. The prepared samples of a rectangular shape were polished as optical-flat for the following optical measurements. The densities of the glass samples were measured by using Archimedes' method at room temperature.

The optical absorption spectra of the glasses were measured in the wavelength range 250-2500 nm using UV-VIS-NIR spectrophotometer (JASCO, V-570) .

3. Results and discussion

3.1. Linear and nonlinear optical properties

The absorption spectrum of 3000ppm Sm_2O_3 doped TNZ glass is shown in Fig. 1. The absorption spectrum is characterized by seven bands centered at 950, 1088, 1238, 1386, 1492, 1530 and 1594 nm, corresponding to the absorptions starting from the ground state $^6H_{5/2}$ to the excited states $^6F_{11/2}$, $^6F_{9/2}$, $^6F_{7/2}$, $^6F_{5/2}$, $^6F_{3/2}$, $^6H_{15/2}$ and $^6F_{1/2}$, respectively. The transitions to energy levels higher than $^6F_{1/2}$ are not observed because of the intrinsic conduction band absorption of the host glasses.

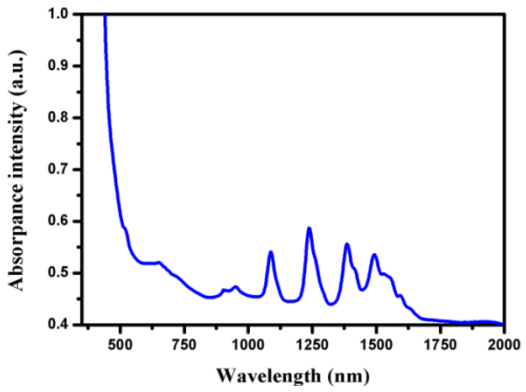

Fig. 1: UV–vis–NIR absorption spectrum of TNZ: Sm_2O_3 glasses.

The linear refractive index depend on the photon energy, $E=h\upsilon$, this can be described by Wemple- DiDomenico relationship [12] :

$$\frac{1}{n^2(\omega)-1} = \frac{E_S}{E_d} - \frac{E^2}{E_S . E_d} \qquad (1)$$

where E_S is the Sellmeier gap Energy and E_d is the dispersion energy, figure (2) shows a plot of $1/n^2(\omega)-1$ versus E^2 for the sample studied TZN-3000ppm Sm. From the linear regression, values of E_s and E_d are obtained (E_s= 6.63 and E_d = 14.9 eV) this value of E_d is higher as compared to the value of E_d for pure SiO_2 and B_2O_3 glass (E_d = 14.71 and 13.2 eV) [13] and the glass system B_2O_3/ PbO/ Al_2O_3/ Sm_2O_3 (E_d =8.49 eV) [14],otherwise, E_d of our sample is lower as compared to the value of E_d for pure crystalline TeO_2 (E_d = 23.2 eV) [15] and the systems of glass TeO_2/ Nb_2O_5 /ZnO/ M_xO_y (where M_xO_y = Ag_2O, PbO and Na_2O) (E_d = 20.6, 21 and 19.78 eV) [16], respectively. The decreasing of E_d may be due to broken bonds (i.e smaller cation coordination

number N_c). Moreover, the decreasing of E_d value may be due to decrease of the covalent bond in the prepared glass sample[17].from the literature it is found that the two parameters E_S and E_d display chemical and structural trends, moreover the parameter E_d is a measure of the strength of interband optical transitions[17].

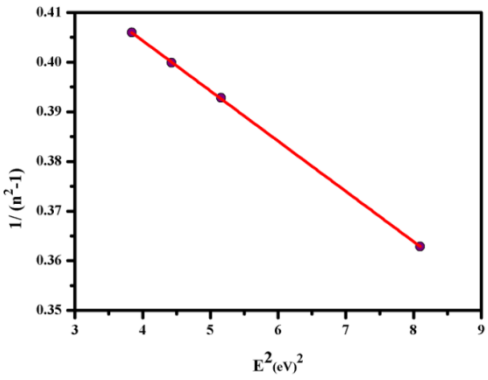

Fig. 2: Refractive indices as a function of the photon energy (illustrated as $1/(n^2-1)$ vs. $(h\upsilon)^2$) of the prepared glass

The refractive index as function of the wavelength this very important parameter for designing of many advanced photonic systems. Moreover, high refractive index increase the optical performance of photonic-crystals through efficient nonlinear interactions [18].The refractive index (n) of the sample has been calculated from the reflectance of the sample at normal incidence by the formula [19] :

$$n = \left(\frac{1+\sqrt{R}}{1-\sqrt{R}} \right) \qquad (2)$$

Figure (3) shows the variation in the refractive index with the incident wavelength. The value of linear refractive index of the prepared glass sample TZN- 3000ppm Sm was found to be 1.858 at 650 nm which is found to be higher than n = 1.698 for the glasses system Li_2CO_3/ B_2O_3/TeO_2/ Sm_2O_3[20], n= 1.79 at 589.3 for the glass system PbO/ TeO_2/ P_2O_5/ ZnO/ $BaCO_3$/ Sm_2O_3 nm[9], n= 1.59 for the glass system PbO/ CaO/ ZnO/ NaF/ B_2O_3/ Sm_2O_3nm [21], n=1.798 at 589.3 nm for the glass system B_2O_3/ PbO/ PbF_2/ Bi_2O_3/ ZnO/ Sm_2O_3[22]. The increase of the refractive index may be due to the increase of the number of non-bridging oxygen (NBO) at the expense of the bridging oxygen (BO) where the (NBO) is more polarizable than the (BO).

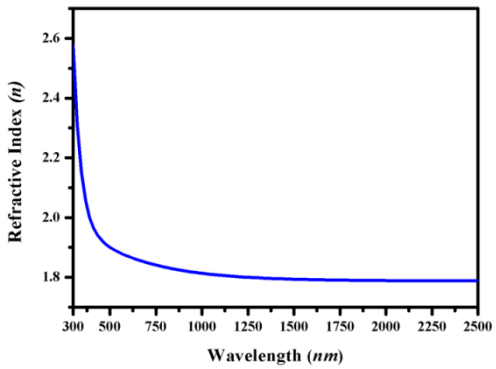

Fig. 3: Refractive index (n) as a function of the wavelength (λ) nm of the prepared glasses TNZ doped with Sm^{3+} ions

Band gap is an important parameter used to analyze the fundamental absorption edge in the UV-region which is a useful method for the investigation of optical transitions and electronic band structure in crystalline and non-crystalline materials. The optical band gap, E_{opt} was calculated using the following general relation proposed for amorphous materials [23] ;

$$\alpha(\omega)=\frac{B\left(\hbar\omega-E_{opt}\right)^{r}}{\hbar\omega} \qquad (3)$$

where, E_{opt}, is the optical band gap energy in eV, *B*, is a constant and the exponent, *r*, is an index which take different values depending on the nature of interband electronic transition responsible for absorption. For glassy materials the allowed indirect transitions are valid according to the Tauc relations [24] and r = 2. $\hbar\omega$ is the photon energy of incident photon. The values of indirect optical band gap energy E_{opt} were determined from equation (3) by the extrapolation of linear region of the $(\alpha\hbar\omega)^{1/2}$ against photon energy $\hbar\omega$ plots at $(\alpha\hbar\omega)^{1/2}$ =0. Fig. (4) show the relation between $(\alpha\hbar\omega)^{1/2}$ and, $\hbar\omega$, for the papered sample, the value of indirect energy gap E_{opt}= 2.38 eV, this value is found to be in the range of other reported systems of glasses likeTeO_2/ Na_2O/ Sm_2O_3/ Yb_2O_3 (E_{opt}=2.73 –2.91eV) [3], PbO/ CaO/ ZnO/ NaF/ B_2O_3/ Sm_2O_3 (E_{opt}= 2.64eV) [21], B_2O_3/ PbO/ PbF_2/ Bi_2O_3/ ZnO/ Sm_2O_3 (E_{opt}= 2.97 –2.94 eV) [22]. Glasses doped with Sm^{3+} ions of large energy gap exhibit high quantum efficiency.

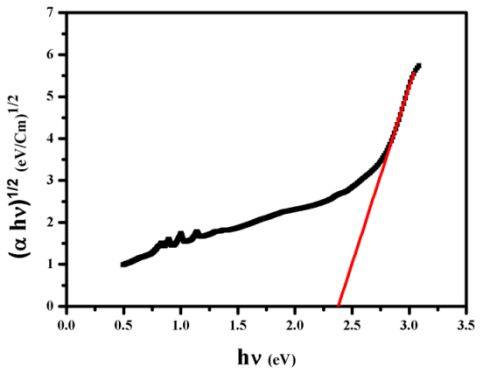

Fig. 4.$(\alpha h\nu)^{1/2}$ as a function of the photon energy, $h\upsilon$, of studied glasses;TNZ-3000 ppm Sm_2O_3

The glasses with large nonlinear optical properties such as (non-linear refractive index n_2, third-order nonlinear optical susceptibility $\chi^{(3)}$and non-linear absorption coefficient, *β)*are promising materials for nonlinear optical devices such as real time holography, ultrafast optical switches, power limiters, self-focusing, white-light continuum generation

The non-linear refractive index of metal oxides was calculated according to the theory of Lines [25, 26] and Kim et al [27];

$$n_2 = L.\frac{(f.f_L)^3.d^2.(n_0^2-1).E_S^6}{n_0\ [E_S^2-E^2]^4} \qquad (cm^3/erg) \qquad (4)$$

where the factor , *f* is the local field enhancement factor , $f_L=(n^2+2)/3$ is the lorentz local field, *d* is the bond length between cation and anion , in angstrom, n_0 is the long-wavelength limiting value of refractive index and E_s is the Sellmeier gap energy and *L* is an empirical factor, *L*=25 x 10^{-13}. The $n_2$value of the prepared glass sample was n_2 =4.83×$\boldsymbol{10^{-15}}$(cm^2/w) , it was summarized in Table (1).this value larger than the value of n_2 for fused silica 0.3×$\boldsymbol{10^{-15}}$(cm^2/w)[28] moreover, it is higher as compared to the systems Borophosphate–30 Nb_2O_5 and Borophosphate–30 TiO_2 where (n_2 =2 and 1.33×$\boldsymbol{10^{-15}}$(cm^2/w), respectively[29, 30].The increase of $n_2$of the prepared glass may be due to the decrease of the energy gap of TeO_2(E_g = 4.49eV) comparing to the energy gap of SiO_2

(E_g = 9.05 eV) and B_2O_3 (E_g = 12.86 eV) [31]. Alls may be due to the increase of cation polarizability of Te^{4+} (1.595 Å^3) comparing with B^{3+} (0.002 Å^3) and Si^{4+} (0.033 Å^3)[32]
In addition, it is in the range of the systemsTeO_2/ WO_3/ Nb_2O_5/ XO where [x=Mg, Zn and Ni] where n_2 =4.47, 4.6 and 4.78 ×10^{-15}(cm^2/w) [28], respectively.

The third-order nonlinear optical susceptibility $\chi^{(3)}$ was calculated from the nonlinear refractive index $n_2$and the linear refractive index n at 800 nm using the following equation

$$\chi^{(3)}\ (esu) = \frac{n}{3\pi} \cdot n_2 \quad (esu) \qquad (5)$$

The value of $\chi^{(3)}$ was 4.1×10^{-13} (esu), it was listed in table (4).this value is larger than that values for pure SiO_2 glass ($\chi^{(3)}$ =2.4 × 10^{-14}esu) and for pure TeO_2 glass ($\chi^{(3)}$= 1.7 × 10^{-13}esu) [33] and also higher than $\chi^{(3)}$ for TeO_2- Nb_2O_5, TeO_2- Nb_2O_5- Na_2O and TeO_2/ WO_3/ Nb_2O_5/ Na_2O (0.94 , 0.49 and 3.82× 10^{-13}esu), respectively[34, 28].

The non-linear absorption coefficient, β can be calculated using the following expression [35, 36]:

$$\beta = \frac{K E_P^{1/2} F}{n^2 E_{opt}^3} \qquad (6)$$

Where K=3100 cm .GW^{-1}, E_p = 21 eV is K an energy parameter, E_{opt} is the energy gap and F is the function which represents the dispersion of β with respect to the incident photon energy $\hbar\omega$. This function depends on the band structure and determines the energy states that are coupled. The function F is calculated from the relation [37, 38];

$$F = \frac{\left[\left(2\hbar\omega / E_{opt}\right) - 1\right]^{3/2}}{\left(2\hbar\omega / E_{opt}\right)^5} \qquad (7)$$

The photon energy $\hbar\omega$ range is selected at wavelength satisfying the two-photon absorption TPA condition that $E_g/2 < \hbar\omega < E_{opt}$. In this work the value of β was 12.07 *(cm/GW)* which is larger than β For pure α-TeO_2 glass (β = 5.12 cm/GW [39, 40] and other many systems such as chalcogenide glass systems Ge-Sb-S-Se (β= 0.1- 4.9 *cm/GW*)[41]. Tellurite glass systems TeO_2 /WO_3 /Nb_2O_5 / M_xO_y where M_xO_y= (Na_2O, Ag_2O, ZnO, MgO, TiO_2) (β= 6.8 – 9.8 *cm/GW*) [28] the increase in the value of β may be due to the decrease in the energy gap where the energy gap of the prepared sample E_{opt}= 2.38 eV is lower as compared to the energy gap of pure α-TeO_2glass (E_{opt}= 4.49 eV) and Tellurite glass systemsTeO_2/ WO_3/ Nb_2O_5/ M_xO_y where M_xO_y= (Na_2O, ZnO, MgO, TiO_2) (E_{opt}= 2.75, 2.53, 2.73 and 2.68 eV), respectively.

3.2.Absorption spectroscopy, emission cross section and gain coefficient

The Judd–Ofelt (JO) theory is widely used for predicting the possibility of laser action, as well as of optical amplification, through an analysis of the forced electric dipole transitions within the $4f^n$ configuration of rare-earth ions in different isotropic lattices (crystalline and amorphous) [25- 27]. The absorption cross-sections of the Sm^{3+} ion for the $^6H_{5/2} \rightarrow {}^6F_{1/2}$ transition can be calculated as follows:

$$\sigma_a(\lambda) = \frac{2.303 \cdot OD(\lambda)}{N L} \qquad (8)$$

Where OD (λ) = log (I_0/I) is the optical density of the experimental absorption spectrum, L is the thickness of the sample and N is the concentration of respective rare-earth ions.

The stimulated emission cross-section $\sigma_e(\lambda)$ of Sm^{3+} for the $^6H_{5/2} \rightarrow {}^6F_{1/2}$transition can be deduced from their corresponding ground state absorption cross-section $\sigma_a(\lambda)$ using the follow equation [28]:

$$\sigma_e(\lambda) = \sigma_a(\lambda) \frac{Z_l}{Z_u} \exp\left[\frac{E_{Zl} - \frac{hc}{\lambda}}{K_B T}\right] \qquad (9)$$

Where Z_l and Z_u are the partition functions for the lower and the upper levels involved in the considered optical transition, T is the temperature (in this case the room temperature), and E_{ZL} is the zero line energy for the transition between the lower Stark sublevels of the emitting multiplets and the lower Stark sublevels of the receiving multiplets.

Fig. (5a) shows the calculated absorption and emission cross sections for the present glasses. The peak of the stimulated emission cross-section (σ_e^{Peak}) is about 1.09 x10^{-20} cm^2 respectively. The highest value of (σ_e^{Peak}) for the emission cross-section is related to the larger value of the line strength of the$^6F_{1/2} \rightarrow {}^6H_{5/2}$and may be due to the high refractive index of the glass matrix. Table 1 shows that the value of the emission cross section at around 1.653μm of the TNZ: Sm glass is larger than those of other glasses. For laser glasses, it is generally desirable for the emission cross section to be as large as possible in order to provide high gain [37]. It indicates that the doped glass TNZ: Sm is a promising candidate for laser glass at 1.653 μm and 1653 nm. The FWHM of the emission peak is also a critical parameter that is used to evaluate the gain bandwidth properties of the optical amplifiers is 45 nm respectively. Due to the large overlap of the absorption and emission spectrum of Sm^{3+} ions at 1.653 μm, re-absorption will occur and cause the fluorescence spectrum deformed. Thus, due to the asymmetric profile of the emission line, it is more reasonable to calculate an effective line width, instead of the FWHM. The effective line width (Δλ) can be expressed as $\Delta\lambda = \int \sigma_e(\lambda)d\lambda / \sigma_e^{Peak}$. The effective bandwidths is 41 nm respectively. In order to understand the band profile of the $^6F_{1/2} \rightarrow {}^6H_{5/2}$emission of the Tm^{3+} ions and estimate the Stark splitting for the $^6F_{1/2}$ emitting and the $^6H_{5/2}$ground levels in the studied tellurite glass, a Gaussian de-convolution of the 1.653 μm band has been performed. Fig. 5b shows the emission spectra due to the $^6F_{1/2} \rightarrow {}^6H_{5/2}$transition of Sm^{3+} ions and the deconvolved Gaussian amplitude peaks obtained from the fitting to the emission spectra obtained for Sm^{3+}-doped (TNZ) glasses (dotted lines). Peak positions and the width of this subcomponent peaks are labeled as A, B and C as shown in Fig. 5b. In order to explain the 1.653 μm emission of the Sm^{3+} ions, an equivalent model of the four levels system is shown in Fig. 5c. The ground $^6H_{5/2}$level splits into two sublevels at around 0 cm^{-1} and 333 cm^{-1}. The excited $^6F_{1/2}$ level also splits into two sublevels (Starks levels) at around 6382.03 cm^{-1} and 6213 cm^{-1} as shown in Fig. 8to gather with all of the transitions possible between these subcomponents. In all samples the energy differences ΔE_1 = 333– 0 = 333 cm^{-1} and ΔE_2 = 6382.03 – 6213 = 169.03 cm^{-1} are the values of the energy range of the Stark splitting of the $^3H_6$ and the $^6F_{1/2}$multiplets, respectively. The ground state presents a larger Stark splitting than the emitting level for the tellurite glass under study, in a similar way as in previous reports on Er^{3+} doped tellurite glasses [35]. The results also indicate that the bandwidth is strongly dependent on the overall extent of the Stark splitting.

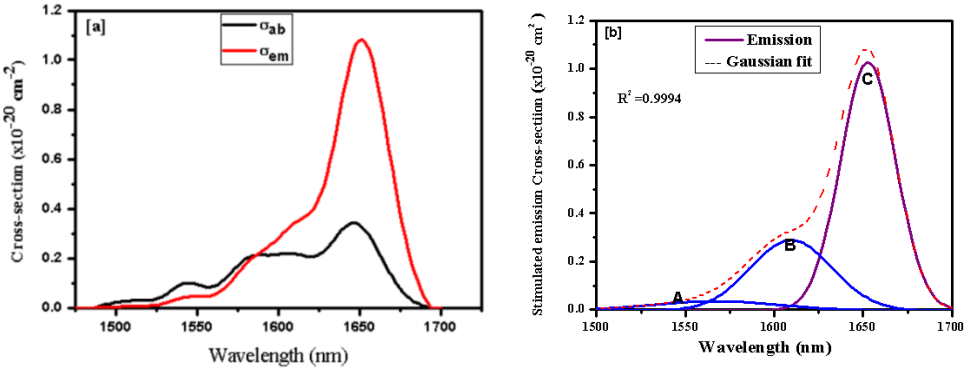

Fig. 5a, b:(a) Absorption$\sigma_a(\lambda)$ and stimulated emission cross sections $\sigma_e(\lambda)$ of the transition $^6H_{5/2} \rightarrow {}^6F_{1/2}$ in TNZ: Sm_2O_3, (b): Deconvolution of emission spectra Gaussian amplitude peaks fitted of TNZ: Sm_2O_3 glass.

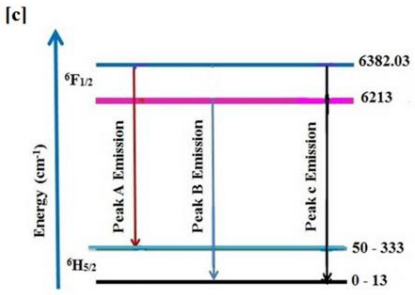

Fig. 5c:An equivalent model of four level system for describing 1.653 µm emission of Sm^{3+} doped present glass TNZ

Optical gain coefficient is an important factor for evaluating the performance of laser media. If P is the population inversion rate for $^6F_{1/2} \rightarrow {}^6H_{5/2}$, Sm^{3+}laser transition, the gain cross section can be calculated using the following relation:

$$\boldsymbol{\sigma}_{gain} = \mathbf{P}\boldsymbol{\sigma}_{em}(\lambda) - (1- \mathbf{P})\, \boldsymbol{\sigma}_{ab}(\lambda) \qquad (10)$$

Where σ_{em} and σ_{abs} are emission and absorption cross section, respectively. The wavelength dependence of the gain cross section was calculated for different values of population inversion P (P = 0, 0.1, 0.2,….. , 1) and are shown in fig. 6.

In the case of total inversion at 1651 nm, a gain coefficient of 1.09 cm^{-1} is obtained for TNZ doped with 3000 ppm Tm_2O_3ions. But, as it is known for such laser systems, the inversion coefficient fraction is more likely close to 0.2 which leads to the gain coefficient values of - 0.09 cm^{-1} at 1640 nm. Laser experiments for the see missions are expected to find light amplification in the future.

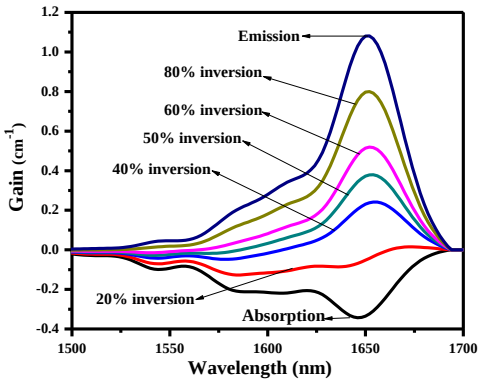

Fig. 6.The gain coefficient of the transition $^6H_{5/2} \rightarrow {}^6F_{1/2}$ for TNZ: Sm_2O_3 glass

4. Conclusions

The luminescence properties of the present glasses with composition $75TeO_2$-$12.5Nb_2O_5$-12.5ZnO doped 3000 ppm Sm_2O_3 were characterized. This results obtain that thesis glasses has optical energy gape E_{opt}= 2.38 eV, Sellmeier energy gap E_s= 6.63 and dispersion energy E_d = 14.9 eV. Moreover it has the third-order nonlinear optical susceptibility $\chi^{(3)}$equal 4.1×10^{-13}(esu) and non-linear absorption coefficient, β , equal 12.07 *(cm/GW)*. The gain cross-section of laser transition level from $^6H_{5/2} \rightarrow {}^6F_{1/2}$was obtained of present glasses and it is (1.09 x 10^{-20} cm^2) finally it has the effective emission cross section bandwidth (41 nm). From these results the present glasses is suitable for using as candidate of optical application.

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