Intersystem crossing effect on photobleaching of $Cr^{+4}:YAG$ saturable absorber as a passive Q-switch

Abdul-Kareem M.Salih
Physics Dep., College of Science , Thi-Qar University, Thi-Qar, Iraq.

Abstract

Mathematical model is presented in this work and solved numerically by Rung- Kutta–Fehelberg to study the effect of molecules intersystem crossing (K_{isc}) on photobleaching (bleaching time) of $Cr^{+4}:YAG$ crystal as a saturable absorber material (passive Q- switch) with $Nd:GdVO_4$ laser. The study show the increasing of molecules intersystem crossing into energy levels decreasing the bleaching time of saturable absorber material to lead fast built-up laser pulse.

***Keywords*: Nonlinear optics absorption - Intersystem crossing $Cr^{+4}:YAG$ crystal**

تأثير العبور البيني في القصر الضوئي للمادة الماصة المشبعة
$Cr^{+4}:YAG$ كمفتاح للتحويل السلبي لعامل النوعية

عبد الكريم مهدي صالح / قسم الفيزياء- كلية العلوم – جامعة ذي قار

الخلاصة

تم صياغة وحل نموذج رياضي باستخدام طريقة رونج – كوته- فهلبرج العددية لدراسة تأثير العبور البيني للجزيئات في المادة الماصة المشبعة ($Cr^{+4}:YAG$) على خاصية القصر الضوئي (زمن القصر) عند استخدام تلك المادة كمفتاح للتحويل السلبي لعامل النوعية مع ليزر ($Nd:GdVO_4$) . بينت الدراسة أن زيادة قيمة العبور البيني للجزيئات بين مستويات الطاقة للمادة الماصة المشبعة يؤدي إلى نقصان زمن القصر أي حدوث حالة الإشباع في الامتصاص بزمن أقل مما يؤدي إلى بناء أسرع لنبضة الليزر.

1. Introduction

Pulsed solid-state laser widely used in scientific, medical, industrial and military systems, the efficiency and cost are very important in this applications. For this, the passive Q-switching has been widely used to get pulsed laser [1,2].The saturable absorber material (S.A.M.) (passive Q- switch) performance depending

on its characteristics such as the energy and lifetime of levels, chemical stability, surface tension, absorption cross section, and optical quality[2-4], then several S.A.M. have been developed to replace the dyes as passive Q- switchs, the most used is undoubtedly $Cr^{+4}:YAG$ crystal (Chromium doped Ytterium Aluminum Garnet), it is an excellent crystal for passive Q-switching in the wavelength range from 800 nm to 1200 nm, because of its a good ratio between its ground and excited levels cross-sections, more over, is optically well known and benefits for excellent optical quality and thermal conductivity [2,5].

The energy levels diagram of $Cr^{+4}:YAG$ crystal is shown in fig. (1) [5,6] the levels $^3A_{2g}$, $^3T_{2g}$ and $^3T_{1g}$ are all spin triplets, whereas ^1E_g and $^1A_{1g}$ are spin singlet levels. In brief, the transition from the ground level $^3A_{2g}$ to the first excited level $^3T_{2g}$ occurs by photons absorption, the optical bleaching at higher fluence occurs when there is an appreciable population in the first excited level $^3T_{2g}$. At $^3T_{2g}$ the excitation to the second excited level $^3T_{1g}$ also occurs by photons absorption due to the $^3T_{2g}$. The decay back to the ground state $^3A_{2g}$ or make a forbidden transition to the ^1E_g state by intersystem crossing mechanism. The effect of intersystem crossing on the nonlinear absorption (photobleaching) of $Cr^{+4}:YAG$ crystal has been studied in this work.

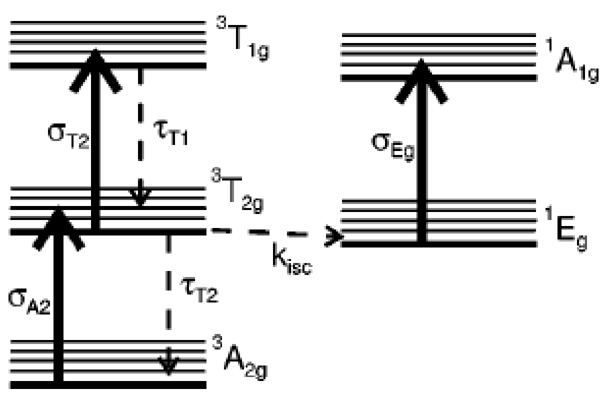

Fig. (1) : Energy levels diagram of $Cr^{+4}:YAG$ crystal

2.Theory

Mathematical model is presented in this work to describe the performance of $Cr^{+4}:YAG$ **passive Q-switch with** $Nd:GdVO_4$ **(Neodymium doped Gadolinium Orthovanadate) laser as the following set of rate equations**

$$\frac{dn}{dt} = (2\sigma_g L_g N_g - 2L_S(\sigma_{A2}N_{A2} + \sigma_{T2}N_{T2} + \sigma_{Eg}N_{Eg}) - (\ln(1/R) + \Gamma))\frac{n}{\tau_R} \quad \textbf{(1-a)}$$

$$\frac{dN_g}{dt} = R_p - \gamma_g N_g - (2\sigma_g L_g / \tau_R)\gamma_p N_g n \quad \textbf{(1-b)}$$

$$\frac{dN_{A2}}{dt} = \gamma_{T2}N_{T2} - (2\sigma_{A2}L_S / \tau_R)N_{A2}n + \gamma_{Eg}N_{Eg} \quad \textbf{(1-c)}$$

$$\frac{dN_{T2}}{dt} = (2\sigma_{A2}L_S / \tau_R)N_{A2}n - \gamma_{T2}N_{T2} - (2\sigma_{T2}L_S / \tau_R)N_{T2}n + \gamma_{T1}N_{T1} - k_{ISC}N_{T2} \quad \textbf{(1-d)}$$

$$\frac{dN_{Eg}}{dt} = K_{isc}N_{T2} + \gamma_{A1}N_{A1} - \gamma_{Eg}N_{Eg} - (2\sigma_{Eg}L_S / \tau_R)N_{Eg}n \quad \textbf{(1-e)}$$

$$\frac{dN_{T1}}{dt} = (2\sigma_{T2}L_S / \tau_R)N_{T2}n - \gamma_{T1}N_{T1} \quad \textbf{(1-f)}$$

$$\frac{dN_{A1}}{dt} = (2\sigma_{Eg}L_S / \tau_R)N_{Eg}n - \gamma_{A1}N_{A1} \quad \textbf{(1-g)}$$

The parameters used in this model are defined as following: n is the photon density in the laser cavity. Ng is the population inversion density of the laser medium , σg is the laser emission cross section, L_g **is the length of the laser gain medium,** L_S **is the length of the S.A.M. crystal,** Γ **is the remaining round-trip cavity dissipation,** τ_R **is the cavity round-trip transit,** $\sigma_{A2}, \sigma_{T2}, \sigma_{Eg}$ **are the absorption cross sections of** $^3A_{2g}$ **level (ground –state),** $^3T_{2g}$ **and** ^1E_g **exited levels of saturable absorber respectively,** N_{A2} **is the population of** $^3A_{2g}$ **level of saturable absorber.** N_{T2} **is the population of** $^3T_{2g}$ **excited level of saturable absorber.** N_{Eg} **is the population of** ^1E_g **excited level of saturable absorber ,** N_{T1} **is the population of** $^3T_{1g}$ **excited level of saturable absorber,** N_{A1} **is the population of** $^1A_{1g}$ **excited level of**

saturable absorber. R_p is the pumping rate. $\gamma_g = 1/\tau_g$, is the decay rate of the upper laser level, τ_g is the upper laser level lifetime. γ_p is the population reduction factor (bottlenecking parameter), γ_p equal 1 for a four level and 2 for three level laser active medium, $\gamma_{T2} = 1/\tau_{T2}$ is the spontaneous decay rate of $^3T_{2g}$ excited level of saturable absorber, τ_{T2} is the lifetime of the $^3T_{2g}$ excited level of saturable absorber. $\gamma_{Eg} = 1/\tau_{Eg}$ is the spontaneous decay rate of ^1E_g excited level of saturable absorber, τ_{Eg} is the lifetime of ^1E_g excited level. $\gamma_{T1} = 1/\tau_{T1}$ is the spontaneous decay rate of $^3T_{1g}$ excited level of saturable absorber, τ_{T1} is the lifetime of $^3T_{1g}$ excited level, $\gamma_{A1} = 1/\tau_{A1}$ is the spontaneous decay rate of $^1A_{1g}$ excited level of saturable absorber, τ_{A1} is the lifetime of $^1A_{1g}$ excited level. K_{isc} is the intersystem crossing from $^3T_{2g}$ into ^1E_g excited levels.

As discussion the set of equations (1); In general, the build-up time of Q-switched laser pulse is very short compared to pumping rate R_p and the relaxation time of active medium τ_g, then it is possible to neglect pumping rate and spontaneous decay of active medium during pulse generation [7,8]. The lifetimes τ_{T2} and τ_{Eg} are much longer than the pulse duration [5], so all the terms with τ_{T2} and τ_{Eg} can be neglected. While the lifetimes τ_{T1} and τ_{A1} are much shorter than pulse duration [5], so all the terms with τ_{T1} and τ_{A1} can be neglected.

According to the previous discussion , the set of rate equations (1) can be written as the following

$$\frac{dn}{dt} = (2\sigma_g L_g N_g - 2L_S(\sigma_{A2}N_{A2} + \sigma_{T2}N_{T2} + \sigma_{Eg}N_{Eg}) - (\ln(1/R) + \Gamma))\frac{n}{\tau_R} \quad \textbf{(2-a)}$$

$$\frac{dN_g}{dt} = -(2\sigma_g L_g / \tau_R)\gamma_p N_g n \quad \textbf{(2-b)}$$

$$\frac{dN_{A2}}{dt} = -(2\sigma_{A2} L_S / \tau_R) N_{A2} n \quad \textbf{(2-c)}$$

$$\frac{dN_{T2}}{dt} = (2\sigma_{A2} L_S / \tau_R) N_{A2} n - k_{ISC} N_{T2} \quad \textbf{(2-d)}$$

$$\frac{dN_{Eg}}{dt} = K_{isc} N_{T2} \quad \textbf{(2-e)}$$

At the onset of Q-switching , can be considering the most population of saturabele absorber material is in the ground state ($^3A_{2g}$ level) $(N_{A2} = N_{ao})$ where N_{ao} is the total number of molecules in saturabele absorber material, also the time variation of the photons density is approximate to zero $(\frac{dn}{dt} \approx 0.0)$. Corresponding to these physical and mathematical approximations can be determined the initial population inversion of active medium by equation (1-a) as the following

$$N_{go} = (2\sigma_{A2} L_S N_{A2} + \ln(1/R) + \Gamma)/2\sigma_g L_g \quad \textbf{(3)}$$

With the continuing pumping and decreasing of saturabele absorber material absorption, the photons density within the cavity increasing rapidly to generate giant laser pulse. Then in the peak of giant pulse, can regard $\frac{dn}{dt} \approx 0.0$, then the threshold value of population inversion (N_{th}) approximate to the following

$$N_{th} = (2\sigma_{T2} L_S N_{T2} + 2\sigma_{Eg} L_S N_{Eg} + \ln(1/R) + \Gamma)/2\sigma_g L_g \quad \textbf{(4)}$$

Where, $N_{T2} = N_{ao} - K_{isc} N_{ao}$ **(5)**

$$N_{Eg} = K_{isc} N_{ao} \quad \textbf{(6)}$$

By dividing equation (2-a) on the equation (2-b) , we get

$$\frac{dn}{dN_g} = (2\sigma_g L_g N_g - 2\sigma_{A2} L_S N_{A2} - 2\sigma_{T2} L_S N_{T2} - 2\sigma_{Eg} L_S N_{Eg} - \ln(1/R) + \Gamma)/(-2\sigma_g L_g \gamma_p N_g)$$

$$\int_{n_i}^{n_p} dn = -\tfrac{1}{\gamma_p}((\int_{N_{g0}}^{N_{th}} dN_g) - ((2L_S(\sigma_{A2} N_{A2} + \sigma_{T2} N_{T2} + \sigma_{Eg} N_{Eg}) + \ln(1/r) + \Gamma)/2\sigma_g L_g) \int_{N_{g0}}^{N_{th}} \tfrac{dN_g}{N_g}) \quad \textbf{(7)}$$

From Eq. (7), the photon number reaches a peak value n_p when population inversion N_g is equivalent to N_{th}, also N_{A2} approaches zero $(N_{A2} \approx 0.0)$, then

$$\int_{n_i}^{n_p} dn = -\tfrac{1}{\gamma_p}(\int_{N_{g0}}^{N_{th}} dN_g - N_{th} \int_{N_{g0}}^{N_{th}} \tfrac{dN_g}{N_g}), \quad \textbf{but } n_p >> n_i \textbf{, then}$$

$$n_p = -\tfrac{1}{\gamma_p}(N_{th} - N_{g0} - N_{th} \ln(\tfrac{N_{th}}{N_{g0}}) \quad \textbf{(8)}$$

After the release of the Q-switched laser pulse, the population inversion is reduced to the final value N_f, this value can be utilized to calculate the output energy of Q-switched pulse using the following equation

$$E_{out} = \left(\frac{N_{g0}-N_f}{\gamma_p}\right)\left(\frac{N_{g0}-N_f}{N_{go}}\right)h\upsilon \quad (9)$$

Where $h\upsilon$ is the laser radiation energy. The photons absorbed by $^3A_{2g}$, $^3T_{2g}$,and ^1E_g levels have been calculated by the following equations respectively

$$Ph..abs.(^3A_2) = \frac{\sigma_{A2} L_S N_{A2}}{\sigma_g L_g} \quad (10)$$

$$Ph..abs.(^3T_2) = \frac{\sigma_{T2} L_S N_{T2}}{\sigma_g L_g} \quad (11)$$

$$Ph..abs.(^1E_g) = \frac{\sigma_{Eg} L_S N_{Eg}}{\sigma_g L_g} \quad (12)$$

3. Numerical simulation

3.1 Calculations

The set of rate equations (2) has been solved numerically by Rung- Kutta – Fehelberg method to study the effect of molecules intersystem crossing on photobleaching (bleaching time) of $Cr^{+4}:YAG$ crystal as a saturable absorber material with $Nd:GdVO_4$ laser. The published values of input data have been used as the following ; from Ref.[5] used the values of $\sigma_{A2} = 5.4\times10^{-18} cm^2$, σ_{T2} = $4\times10^{-18} Cm^2$, K_{isc} = $2-3\times10^8 Sec^{-1}$, while from Ref. [6] used the values of λ =1064 nm, L_c =60 mm, Laser rod dimension= $3.5\times3.5\times4 mm^3$, $\sigma_g = 7.6\times10^{-19} cm^2$, τ = $90\mu s$, S.A.M dimension= $5\times5\times1.74 mm^3$, $N_{ao} = 2.7\times10^{17} cm^{-3}$, Γ =0.2,R=0.75. σ_{Eg} = $3\times10^{-19} cm^2$ [5,6], γ_p =1 [2,5,6].

3.2 Results and Discussion

The study show; the increasing of S.A.M. molecules intersystem crossing into energy levels lead to decreasing of bleaching time of S.A.M., this means the required time in order saturation of nonlinear absorption (photobleaching) will be decreasing , this behaviour shown in fig.(2) and fig.(3). Fig. (3) show the max photon density of passive Q-switching pulses reaches at shorter times while increasing intersystem crossing. The study explain this result, because of

intersystem crossing, the molecules population in $^{1}E_g$ level will be increasing, while the molecules population in $^{3}T_{2g}$ level decreasing in same population of $^{1}E_g$ level increasing. Because of these levels have different absorption cross section , the decreasing of absorption activity of $^{3}T_{2g}$ level have the dominator passively effect more than $^{1}E_g$ level absorption activity increasing in the total (global) absorption activity of S.A.M.. Fig. (4-a) show the absorption due the levels with out intersystem crossing. Figs. (4-b,c,d) show the reduction of photons which are absorbed by $^{3}A_{2g}$, $^{3}T_{2g}$ levels and the increase of photons absorbed by $^{1}E_g$ level when the intersystem crossing increase. Fig. (5) show the profile of the total photons absorbed in S.A.M. as a function of intersystem crossing, it is clear that the saturation state of total nonlinear absorption occur faster (shorter time) when the intersystem crossing increasing which is lead to high photon oscillation in the cavity and rapidly decreasing in population inversion of active media (shown in fig.(6)) to generate fast built-up laser giant pulses (shown in fig.(3)) with little decreasing in energy because of the little increasing in final values of population inversion of active medium when intersystem crossing increasing (shown in fig.(7)).

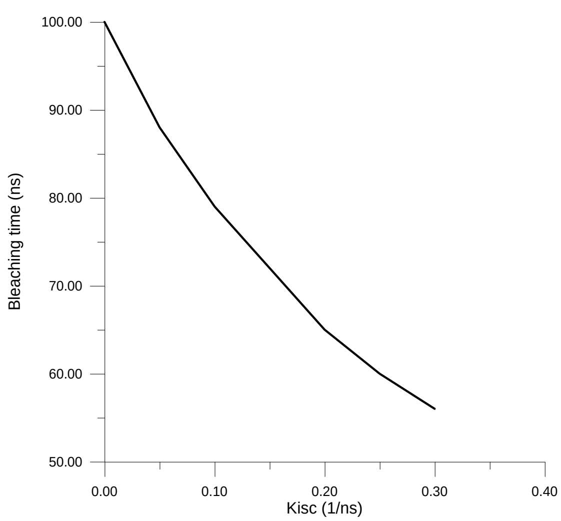

Fig.(2): The variation of bleaching time as a function of intersystem crossing .

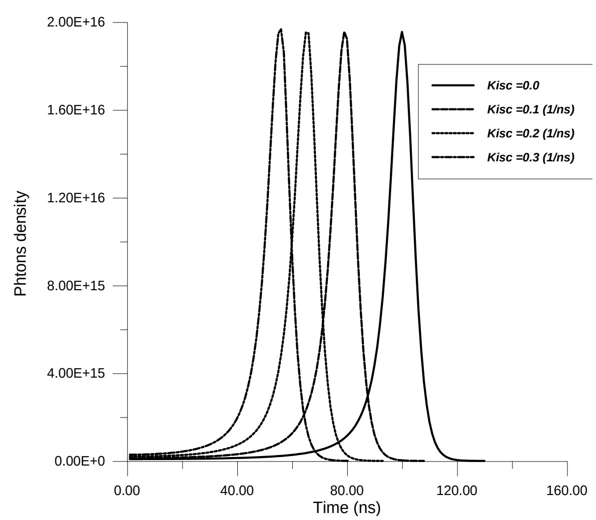

Fig.(3): The profiles of giant pulse related to deferent K_{isc} values.

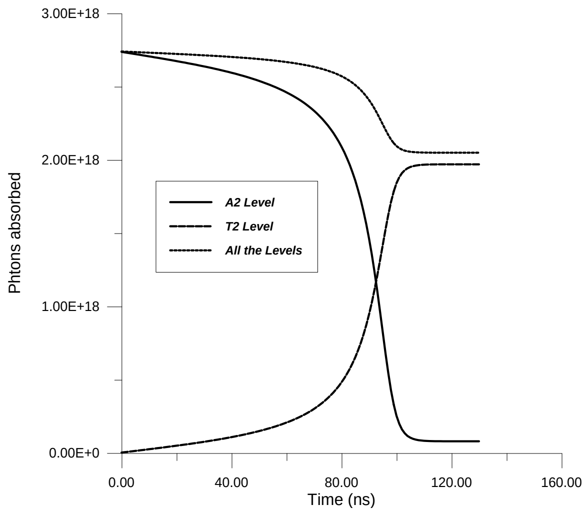

Fig. (4-a)

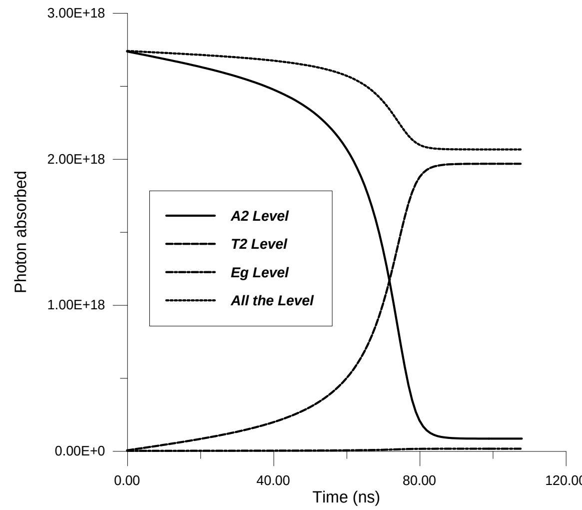

Fig. (4-b)

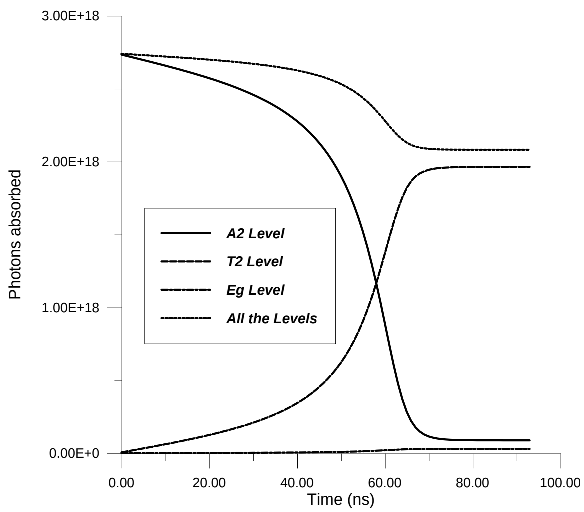

Fig. (4-c)

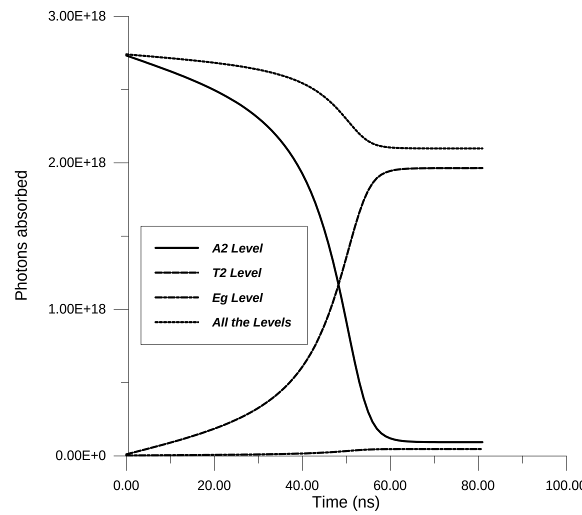

Fig. (4-d)

Fig. (4) : show the photons absorbed by the energy levels of S.A.M. :

(4-a) K_{isc}=0.0, **(4-b)** $K_{isc} = 1\times10^{8}(1/\sec)$, **(4-c)** $K_{isc} = 2\times10^{8}(1/\sec)$,

(4-d) $K_{isc} = 3\times10^{8}(1/\sec)$.

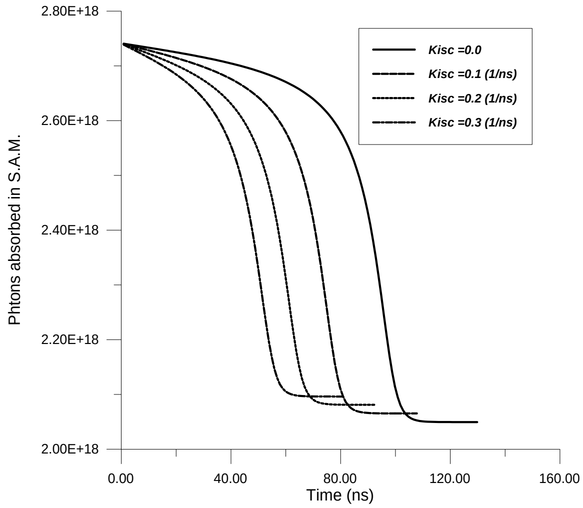

Fig. (5) : The total photons absorbed in S.A.M. as a function of intersystem crossing (K_{isc}) .

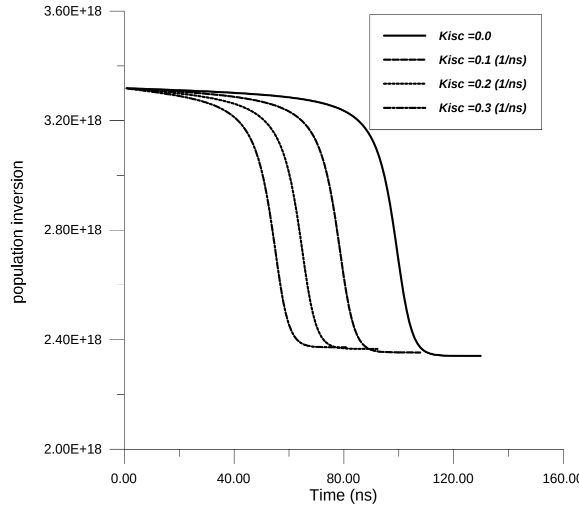

Fig. (6) : Population inversion in active medium as a function of intersystem crossing (K_{isc}).

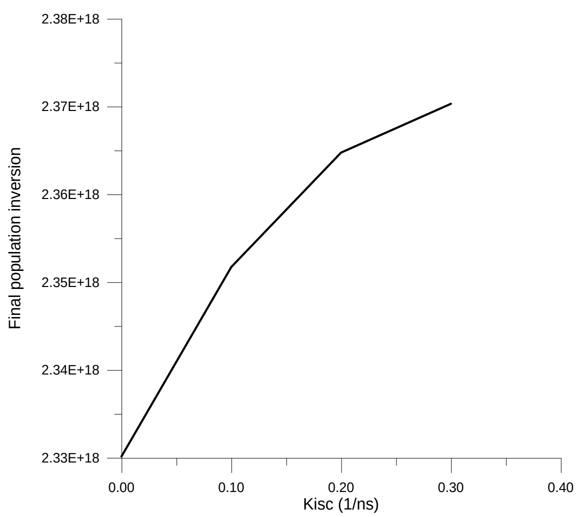

Fig. (7) : Final population inversion of active medium as a function of intersystem crossing

References

[1]- Yen-Kuang, Kuo and Yi-An Chang," Numerical study of passive Q- switching of a Tm: YAG laser with a Ho:YLF solid-state saturable absorber ", Applied Optic,Vol.42, No.9,PP.(1685-1691**), (2003).**

[2]- S. Sebastien, F. Druon, F. Balembois et al., "Passively Q-switched diode -pumped $Cr^{3+}:YAG/GdVO_4$ monolithic microchip laser", Optics communications, 259, P(816-819), **(2005).**

[3]- Abdul-Munim K. Al-Kamil, Abdul-Kareem M. Salih, "Solvent surface tension effect on passive liquid Q- switch pulse duration " J. of Thi-Qar Science, V(1), No.(1), P(73), **(2008).**

[4]- Fanting Kong, Liping Liu, Charlotte Sanders, and Y.C. Chen," Phase locking of nanosecond pulse in a passively Q- switched two –element fiber laser array", Appl. Phys. Lett., V (90), 151110, **(2007).**

[5]-Sang-Hoon Yim, D0ng-Ryeol Lee, Bum Ku Rhee, and doseok," Nonlinear absorption of $Cr^{3+}:YAG$ studied with lasers of different pulse widths", Applied Physics Letters, V(73), No (22), P(3193), **(1998).**

[6]- Iftitan M. Azzouz and Adel El-Nozahy," Numerical study of $Cr^{3+}:YAG$ passively Q-switching $Nd:GdVO_4$ laser ", Egept J. Solids, V(29), No. (1), P(215), (2005).

[7]- A.E.Siwgman," Lasers ", University Science, Millvally, California, P. (1024), **(1986).**

[8] – Abdul-Kareem M. Salih. ," Photons Loss Simulation during Cr:YSO Passive Q-switched Ruby Laser Pulse Generation" Jornal of Education College (Al-Mustanseriya University) , V(3), P.(688), **(2008). .**