Numerical analysis of molecules intersystem crossing effect on a passively Q- switched laser pulse characteristics

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Abstract

The effect of molecules intersystem crossing (K_{isc}) on characteristics (energy and duration) of a Passive Q- switched Laser Pulse has been studied by mathematical description (rate equations model) for temporal performance of which was used as a saturable absorber material (passive switch) with laser. The study shows that the energy and duration pulse are decreasing while the molecules intersystem crossing into saturable absorber energy levels is increasing.

Key words
Laser,
Passive Q-switching,
Intersystem crossing.

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تحليل عددي لتأثير العبور البيني للجزيئات على خصائص نبضة الليزر عند التحويل السلبي لعامل النوعية

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الخلاصة

درس تأثير العبور البيني لجزيئات المادة الماصة القابلة للتشبع على طاقة و أمد نبضة الليزر عند التحويل السلبي لعامل النوعية من خلال الحل العددي لمعادلات المعدل التي تحكم التصرف الزمني لبلورة $Cr^{+4}:YAG$ كمادة ماصة قابلة للتشبع (مفتاح التحول السلبي) مع ليزر $Nd:GdVO_4$. وقد خلصت الدراسة إلى حصول نقصان في قيمة طاقة وأمد النبضة مع زيادة العبور البيني للجزيئات.

Introduction

Pulsed solid-state laser widely used in scientific, medical, industrial and military systems, the efficiency and the cost are very important in these 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 depends 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 good ratio between its ground and excited levels cross-sections, moreover, its optically well known and has benefits for excellent optical quality and thermal conductivity [2,5].

The energy levels diagram of $Cr^{+4}:YAG$ 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 $^3T_{2g}$ is solely responsible for the linear, and the optical bleaching at higher fluence which

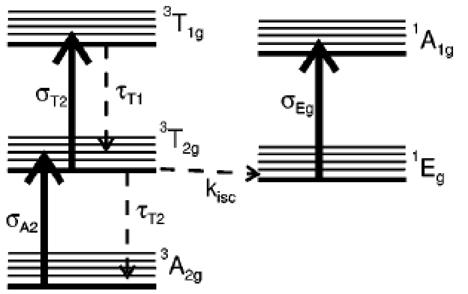

***Fig.(1): The energy levels diagram of** $Cr^{+4}:YAG$*

At $^3T_{2g}$, the excitation to the second excited level $^3T_{1g}$ also occurs by photons absorption by the $^3T_{2g}$. The decay goes back to the ground state $^3A_{2g}$ or makes a (forbidden) transition to the ^1E_g state by intersystem crossing mechanism. The effect of molecules intersystem crossing (K_{isc}) on characteristics (energy and duration) of a Passive Q- switched Laser Pulse has been investigated theoretically in this work for $Cr^{+4}:YAG$ which was used as a saturable absorber material (passive switch) with $Nd:GdVO_4$ laser.

Theory

The performance of $Cr^{+4}:YAG$ as a passive Q-switch with $Nd:GdVO_4$ (Neodymium doped Gadolinium Orthovanadate) laser has been described by the following set of rate equations [7].

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

$$\left[\frac{dN_g}{dt} = R_p - \gamma_g N_g - (2\sigma_g L_g/\tau_R)\gamma_p N_g n\right] \qquad (1\text{-b})$$

occurs when there is an appreciable population in the first excited level $^3T_{2g}$ [5].

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

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

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

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

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

The parameters used are defined as follows: n is the photons number in the laser cavity. Ng is the population inversion density of the laser medium , σ_g 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, τ_R is the cavity round-trip transit, σ_{A2} is the absorption cross section of the level $^3A_{2g}$ (ground –state) of saturable absorber. σ_{T2} and σ_{Eg} are the absorption cross section of the levels $^3T_{2g}$ and ^1E_g respectively, N_{A2} is the population of the level $^3A_{2g}$. N_{T2} is the population of the excited level $^3T_{2g}$. N_{Eg} is the population of the excited level ^1E_g , N_{T1} is the population of the excited level $^3T_{1g}$, N_{A1} is the population of excited level $^1A_{1g}$. 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 the excited level $^3T_{2g}$ of saturable absorber, τ_{T2} is the lifetime of the excited level $^3T_{2g}$ of saturable absorber.

$\gamma_{Eg} = 1/\tau_{Eg}$ is the spontaneous decay rate of the excited level ^1E_g of saturable absorber, τ_{Eg} is the lifetime of the excited level ^1E_g. $\gamma_{T1} = 1/\tau_{T1}$ is the spontaneous decay rate of the excited level $^3T_{1g}$, τ_{T1} is the lifetime of the excited level $^3T_{1g}$, $\gamma_{A1} = 1/\tau_{A1}$ is the spontaneous decay rate of the excited level $^1A_{1g}$, τ_{A1} is the lifetime of the excited level $^1A_{1g}$. K_{isc} is the intersystem crossing from excited level $^3T_{2g}$ into excited level ^1E_g.

The build-up time of Q-switched laser pulse is very short compared to pumping rate R_p and the relaxation time of gain medium τ_g, then it is reasonable to neglect pumping and spontaneous decay of the laser population inversion during pulse generation [8] (the first and the second terms in Eq.(1-b) are neglected). The lifetimes τ_{T2} and τ_{Eg} are much longer than the timescale considered, while τ_{T1} and τ_{A1} are much shorter than the timescale considered [5] (the first and the third terms in Eq.(1-c), the second, third, and the fourth terms in Eq.(1-d), and the second, third, and the fourth terms in Eq.(1-e) are all neglected) . Also the contribution of Eqs.(1-f,1-g) in simulation is neglected.

According to the previous physical conditions, the set of rate Eq.(1) can be written as the following

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

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

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

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

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

At the onset of Q-switching, most population of the saturable absorber material can be considered in the ground state $(^3A_{2g})(N_{A2} = N_{ao})$ 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, the initial population inversion of active medium can be determined by equation (1-a) as the following

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

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

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

where,

$$N_{T2} = N_{ao} - K_{isc}N_{ao} \quad (5)$$

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

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

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

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

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

$$\int_{n_i}^{n_p} dn = -\tfrac{1}{\gamma_p}(\int_{N_{g0}}^{N_{th}} dN_g - N_{th}\int_{N_{g0}}^{N_{th}} \frac{dN_g}{N_g})$$

but $n_p >> n_i$, then

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

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

$$E_{out} = (\tfrac{N_{g0}-N_f}{\gamma_p})(\tfrac{N_{g0}-N_f}{Ngo})h\upsilon \quad (9)$$

where $h\upsilon$ is the laser radiation energy.

Numerical Simulation Calculations

A computer program has been prepared in this study to solve the set of rate equations (2) numerically by Rung-Kutta –Fehelberg method to study the effect of molecules intersystem crossing (K_{isc}) on the characteristics of a Passive Q- switched Laser Pulse. $Cr^{+4}:YAG$ was used as a saturable absorber material (passive switch) with $Nd:GdVO_4$ laser. The published values of input data have been used are shown in Table (I).

Results and Discussion

The study shows the increasing of molecules intersystem crossing into saturable absorber energy levels leads to decreasing the energy and the duration of Q- switched laser pulse as shown in Figs.(2 and 3) respectively. Concerning Fig.(2) ; the study relates this result to the increasing of photons loss which occurs due to the increase of absorption activity of saturable absorber while molecules intersystem crossing is increasing as shown in Figs.(4 and 5), Fig.(4) shows the temporal behavior of photons loss, it is

Parameter	Value	Reference
σ_{A2}	$5.4\times10^{-18} cm^2$	[5]
σ_{T2}	$4\times10^{-18} Cm^2$	[5]
K_{isc}	$2-3\times10^8 Sec^{-1}$	[5]
λ	1064 nm	[6]
L_c	60 mm	[6]
Laser rod dimensions	$3.5\times3.5\times4 mm^3$	[6]
σ_g	$7.6\times10^{-19} cm^2$	[6]
τ	$90 \mu s$	[6]
S.A.M Dimension	$5\times5\times1.74 mm^3$	[6]
N_{ao}	$2.7\times10^{17} cm^{-3}$	[6]
Γ	0.2	[6]
R	0.75	[6]
σ_{Eg}	$3\times10^{-19} cm^2$	[5,6]
γ_p	1	[2,5,6].

clear that the increasing of photons loss with molecules intersystem crossing is

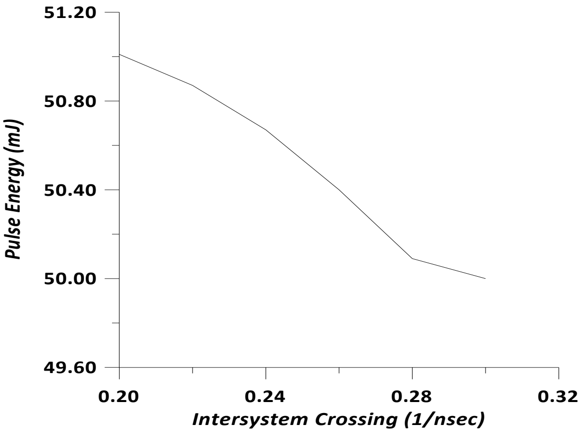

Fig.(2): The variation of pulse energy with intersystem crossing

increasing, Fig.(5) enhances the conclusion of Fig.(4), it shows the increasing of the final values of photons loss via molecules intersystem crossing increasing which is reduces the photons oscillation in the cavity to diminish the photons stimulated emission to cause accumulation in the population inversion which appears in Fig. (6), which shows the increasing of final values of population inversion versus increasing of molecules intersystem crossing increasing, that mean a little amount of energy remains conserved in the active medium instead of released as a laser pulse photons (shown in (Fig.(7)).

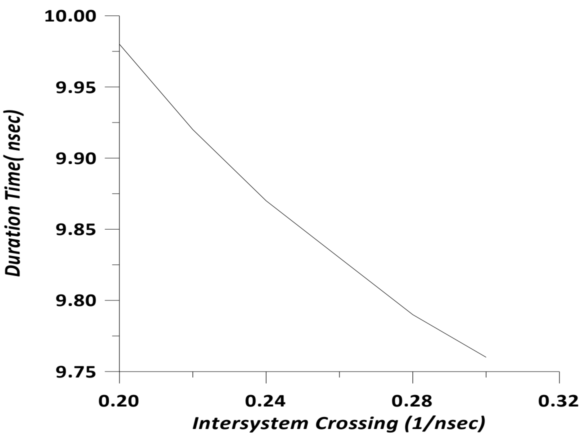

Fig.(3): The variation of pulse duration with intersystem crossing

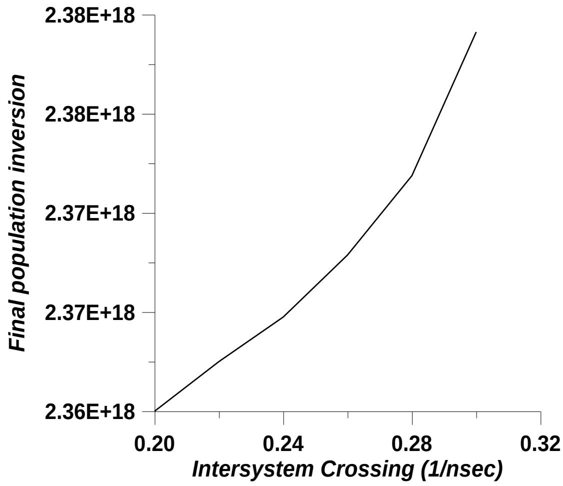

Fig.(4): The profile of photons loss as a function of time

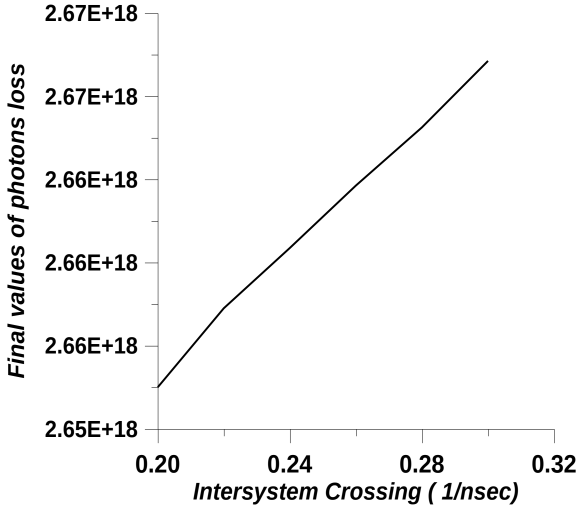

Fig.(5): The final values of photons loss as a function of intersystem crossing

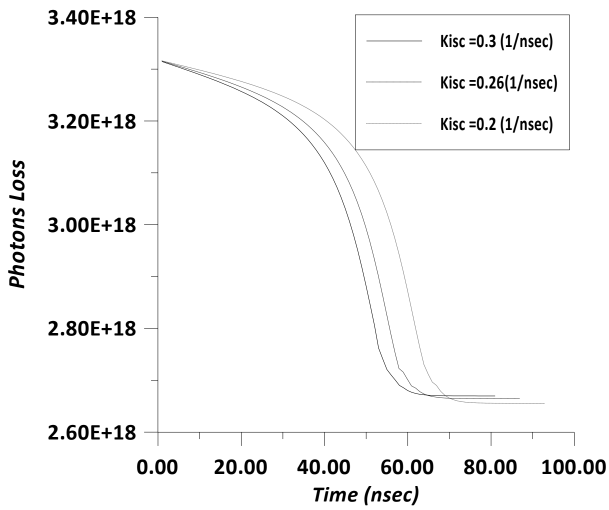

Fig.(6): The final values of population inversion as a function of intersystem crossing

Fig.(7) shows the profile of photon number of passive Q-switching pulse, there is a little decrease which occur while molecules intersystem crossing is increasing ($1.96703\times10^{16} at K_{ISC}=0.2 n\sec^{-1}$) and ($1.952452\times10^{16} at K_{ISC}=0.3 n\sec^{-1}$). Concerning the reduction of pulse duration versus increase of molecules intersystem crossing increasing(shown in Fig.(3)), the study gives an interpretation, because of the amount of energy remains conserved in active medium instead of utilization in passive Q-switching pulse built-up, this status leads to faster falling time of pulse to occasional reduction in pulse duration as shown in Fig. (8).

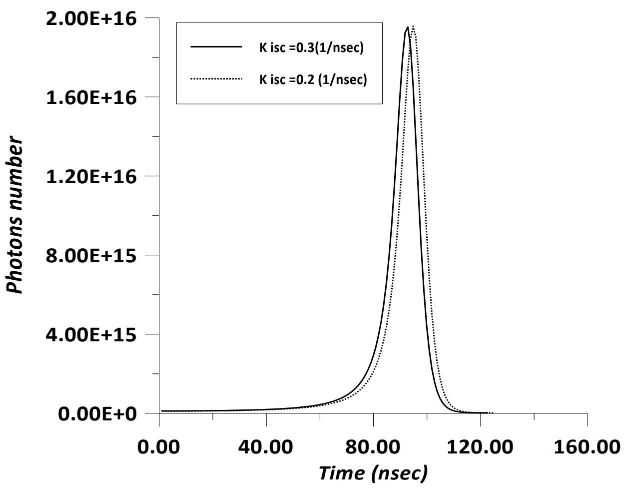

Fig.(7): The profile of photons number as a function of time

Conclusions

The duration and the energy of passive Q-switching pulse are decreasing with the increase of intersystem crossing of saturable absorber material molecules.

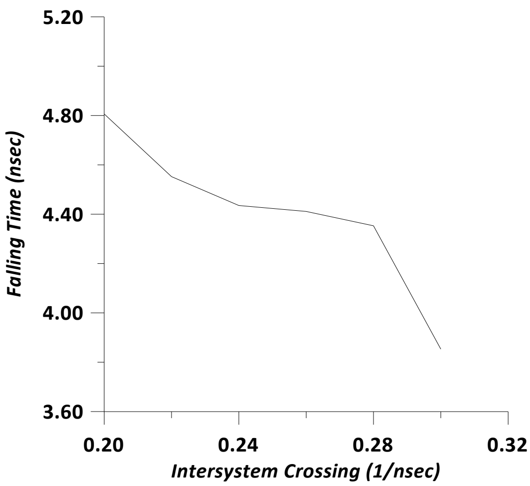

Fig.(8): The falling time as a function of intersystem crossing

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