RP Current Trends in Engineering and Technology
Vol. 1, No. 3 (September 2022) pp. 46–49
e-ISSN: 2583-5491

RESEARCH PLATEAU PUBLISHERS

Cite this article: N. Singh, S. Dahiya, Optimization of all-optical light modulation in nonlinear molecular absorbers, *RP Cur. Tr. Eng. Tech.* **1** (2022) 46–49.

Original Research Article

Optimization of all-optical light modulation in nonlinear molecular absorbers

Ritu*
Department of Basic and Applied Sciences, Bhagat Phool Singh Mahila Vishwavidyalaya, Khanpur Kalan – 131305, India
*Corresponding author, E-mail: ritujakhar42@gmail.com

ARTICLE HISTORY
Received: 07 July 2022
Revised: 03 Sept. 2022
Accepted: 05 Sept. 2022
Published online: 05 Sept. 2022

KEYWORDS
Optical light modulation; nonlinear molecular absorbers; small signal absorption coefficient; saturable absorption.

ABSTRACT
In this study, optical light modulation in nonlinear molecular absorbers is optimised. For nonlinear absorbing thin materials, we were able to derive the general expression for the ideal value of the normalised small-signal absorption coefficient at probe wavelength. We use the complex photocycle of all-optical light modulation in bR as an illustration. Additionally, we thoroughly examined how the optimal value of the normalised small-signal absorption coefficient is affected by the conversion between reverse saturable absorption and saturable absorption in bR. It has also been investigated how different spectral, kinetic, and pump and probe intensities affect the optimal normalised small-signal absorption coefficient value for maximal modulation. The ideal value of the normalised small-signal absorption coefficient is demonstrated to decrease. It's interesting to note that the highest modulation with pump intensity, the best value for the normalised small-signal absorption coefficient, and the corresponding variations of normalised transmitted probe intensity all follow the same pattern. Designing all-optical devices based on nonlinear absorption would benefit from the analysis.

1. Introduction

In case of optical information processing systems, optical control is crucial [1, 2]. In designing of light modulators for broad-band and ultra-fast information processing is the subject of intense research right now [2]. The process of excited-state absorption (ESA), where an intense light signal stimulates molecules from ground to excited-state and exchanges the propagation of the probe signal is a promising method in case of optical modulations [3-6]. In comparison to other techniques, such as optical phase reversal and interference processes, all-optical light modulation dependent on ESA utilizing the pump-probe technique is the straightforward, adaptable, and practical methodology for advantageous purposes. Different all-optical devices, such as optical modulators, optical switches, photonic logic gates, active and passive limiters, bistable devices, etc., can be realised using the fundamental all-optical light modulation process [3-6]. These devices' performance can be accessed via depth of optical modulation, recovery time, power dissipation, and other factors.

To improve the performance of ESA-based all-optical systems, there have been significant research efforts to optimise external parameters including control beam intensity and transverse and longitudinal concentration profile [7-9]. The creation of a generic theoretical framework that could be used to any nonlinear absorbers and the expression of optimization in terms of a normalised parameter would both be advantageous. In our earlier research, we stated that there is a maximum value of normalised small-signal absorption coefficient $\beta = NL\sigma_{1p}$, for which the probe beam can be modulated to its fullest extent [4, 5], where N is the molecular density, L is the sample's size, and σ_{1p} is the cross-section of corresponding to loss (absorption) of lowest stage.

We provide a generic analytical expression for the ideal value of $\beta(\beta_{opt})$ for thin nonlinear absorbing substances in this study. We take the optical modulation in the bacteriorhodopsin (bR) protein as an example since it exhibits a complicated photocycle and has gotten the greatest attention for a variety of bio-molecular photonic applications because of its distinct benefits [10, 11]. We have also analysed the effect of the conversion between saturable absorption (SA) and reverse saturable absorption (RSA) in bR on β_{opt}. An impact of numerous parameters and pump and probe intensity has been studied in detail on β_{opt} for maximum modulation ($\eta_{\max}$). The analysis would be useful to design nonlinear absorption dependent optoelectronic devices.

2. Theoretical model

In the pump-probe approach, the intensity-generated population transformations in nonlinear optical material caused by a single or multiple pump beams can modulate the transmittance of the probe signal when it passes through the material [11, 12]. The expression for the nonlinear intensity-generated coefficient of absorption corresponding to the excited-state may be expressed as:

$$\alpha_p(I_m) = \sum_{i=1}^{n} N_i \sigma_{ip} ,$$

where N_i represents concentration of i^{th} state, σ_{ip} represents the absorption cross-section of the corresponding state at probe wavelength, and I_m is photon density flux (defined as the ratio of intensity of optical signal to the photon's energy) of pump

beam. The total number of molecules in *n* different states are:

$$N = \sum_{i=1}^{n} N_i \, .$$

Consequently, the following is a textual representation of the intensity-induced excited-state absorption coefficient at probing wavelength:

$$\alpha_p(I_m) = N\sigma_{1p} \frac{\sum_{i=1}^{n} N_i \sigma_{ip}}{\sum_{i=1}^{n} (N_i / N_1)} \, .$$

It is possible to express the normalised transmitted probe intensity (NTPI) through the L-thick nonlinear optical material of the sample, modified by the intensity of the pump beam, as follows:

$$\frac{I_{p,out}}{I_{p,in}} = \exp[-\alpha_p(I_m)L] \, .$$

As a result, the NTPI can be expressed as follows via optical signal generated carrier concentrations of various intermediates:

$$\frac{I_{p,out}}{I_{p,in}} = \exp\left[-\beta \frac{\sum_{i=1}^{n} N_i \sigma_{ip}}{\sum_{i=1}^{n} (N_i / N_1)}\right],$$

where $\beta(= N\sigma_{1p}L)$ is the probe wavelength's normalised small-signal absorption coefficient [11, 12]. Using the rate equation method, the optical signal generated carrier concentrations of different states may be described in terms of pump intensity and kinetic and spectral properties of the material. The NTPI can be expressed generally as:

$$\frac{I_{p,out}}{I_{p,in}} = \exp[-\beta f(I_m)] \, ,$$

where $f(I_m)$ is a function of pump intensity.

The modulation of the probe signal intensity between pumping intensities I_{m1} and I_{m2} is the difference in the probe beam's transmission when these two pump intensities are present, and it is stated as:

$$\eta = \left|\exp[-\beta f(I_{m1})] - \exp[-\beta f(I_{m2})]\right| \, .$$

Applying the maxima condition to obtain the ideal value of will allow you to compute the maximum modulation β of probe signal for specific pumping intensities as follows:

$$\beta_{opt} = \frac{\ln[f(I_{m2}) / f(I_{m1})]}{(I_{m2}) - f(I_{m1})} \, . \qquad (1)$$

As a result, the maximum probe beam modulation can be determined by:

$$\eta_{\max} = \left|\exp[-\beta_{opt} f(I_{m1})] - \exp[-\beta_{opt} f(I_{m2})]\right| \, .$$

These are the general expressions of β_{opt} and $\eta_{\max}$ and can be calculated for any molecular configuration by calculating the function $f(I_m)$. The best value of β is owing to a nonlinear fluctuation in probe beam transmission with pump intensity. By using the classic example of rhodopsin proteins, which due to their intricate photocycle incorporate nearly all potential nonlinear absorption processes, the physical mechanism is clarified [11, 12].

Halobacterium halobium purple membrane fragments, which have become an attractive material for bio-molecular photonic applications, have undergone a thorough research of optimization for bR [10–12]. The function $f(I_m)$ has been assessed for the condensed version of the bR typical photocycle [11, 12]. The wild-type bR molecule goes through a number of structural changes as a result of absorbing green-yellow light, leading to the generation of many transitional stages, including B→K→L→M→N→O→B [12]. Rate equations can be used to calculate the carrier concentrations of various stages via excitation routes [12]. Utilizing the steady-state intensity-generated carrier concentration of a variety of states, the function $f(I_m)$ for bR can be estimated and stated as follows:

$$f_{bR}(I_m) = \frac{1 + \sum_{j=2}^{6} I_m \sigma_1 \psi_{12} \left(\frac{\prod_{u=2}^{j-1} k_u}{\prod_{u=2}^{j} (k_u + I_m \sigma_u)} \right) \left(\frac{\sigma_{jp}}{\sigma_{1p}} \right)}{1 + \sum_{j=2}^{6} I_m \sigma_1 \psi_{12} \left(\frac{\prod_{u=2}^{j-1} k_u}{\prod_{u=2}^{j} (k_u + I_m \sigma_u)} \right)} \, . \qquad (2)$$

where N_i (i – 1 to 6) represent the carrier concentrations of B, K, L, M, N and O states in bR photocycle respectively; σ_i and k_i stand for the absorption cross-sections and rate constants of states; $\psi_{12} = 0.64$ is the quantum efficiency corresponding to transition B→K in bR [12], and I_m represent the density of photon flux of the modulated optical signal.

Table 1. Rate constants and absorption cross-sections of bR [12, 13].

Rate constant	Value (s^{-1})	Absorption cross-section	Calculated value (cm^2)	
			570 nm	640 nm
		σ_1	2.4×10^{-16}	3.8×10^{-17}
k_2	3.9×10^{5}	σ_2	1.8×10^{-16}	1.3×10^{-16}
k_3	1.3×10^{5}	σ_3	1.5×10^{-16}	2.7×10^{-17}
k_4	3.8×10^{3}	σ_4	0	0
k_5	2.5×10^{2}	σ_5	1.7×10^{-16}	3.8×10^{-17}
k_6	1.3×10^{2}	σ_6	1.1×10^{-16}	2.5×10^{-16}

3. Results and discussions

By using bR as a typical example, the analysis for optimizing of β in nonlinear absorbers has been conducted. Using Eq. (2) and taking into account the six intermediates in its typical photocycle and the material parameters listed in Table 1, the optical modulational features of bR, specifically the variation of NTPI at the wavelength of 640 nm, corresponding to the peak absorption wavelength of O-state, and modulating optical intensity ($I_m^{'}$) at the wavelength of 570 nm, corresponding to the peak absorption wavelength of B-state, have been simulated [12, 13].

As seen in Figure 1(a), the linear NTPI is high in non-appearance of the optical signal because an initial B-state alone exhibits a low linear probe beam absorption. The NTPI first declines with increasing $I_m^{'}$ values due to greater probe beam absorption via populated O-state, and for larger $I_m^{'}$ magnitudes, the NTPI begins to increase and eventually reaches saturation due to increased optical signal absorption via O-state [12]. So, at smaller and larger optical intensities, the probe beam at 640 nm exhibits reverse-saturable and saturable absorption, respectively.

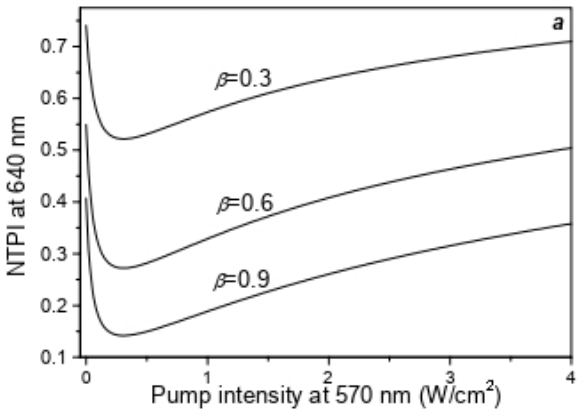

Figure 1. Nature of the dependence of NTPI (at the wavelength of 640 nm) on the pump intensity $I_m^{'}$ (at the wavelength of 570 nm) for three values of probe wavelength's normalised small-signal absorption coefficient β.

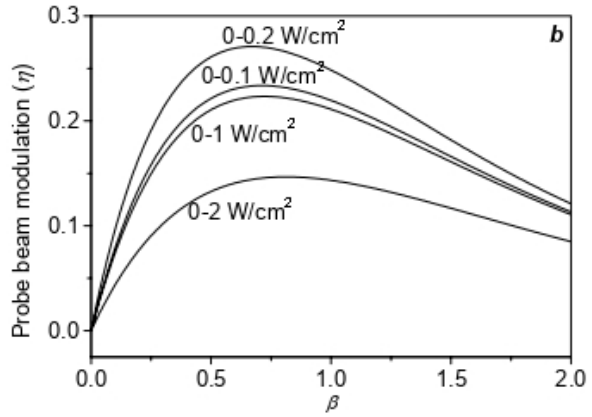

Figure 2. Variation of modulation of NTPI between pump intensities 0 and $I_m^{'}$, with β for different $I_m^{'}$ values.

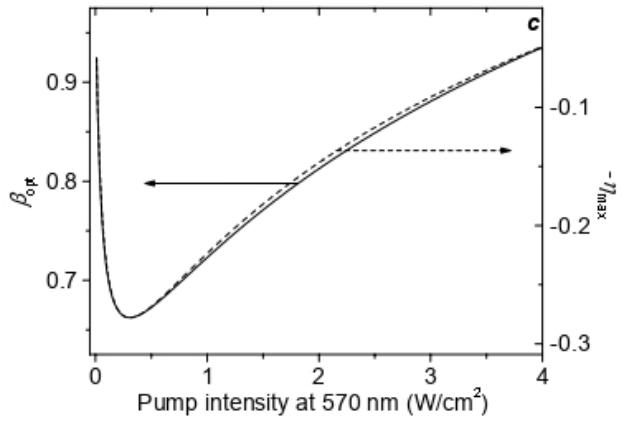

Figure 3. Variation of optimum β for maximum modulation of NTPI between pump intensities 0 and $I_m^{'}$ and negative value of its corresponding maximum modulation, with $I_m^{'}$ at 570 nm.

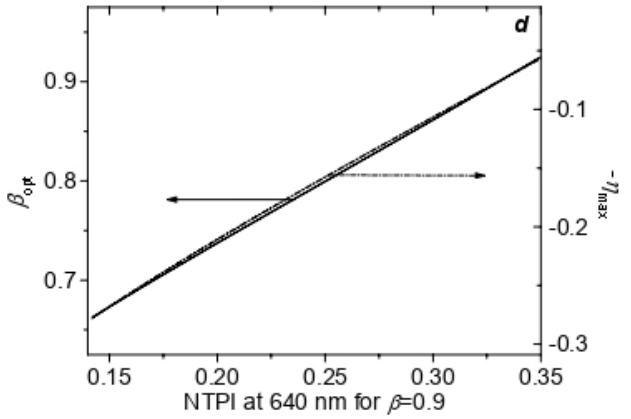

Figure 4. Parametric variation of optimum β and its corresponding negative maximum modulation with NTPI at 640 nm for β =0.9 for a given $I_m^{'}$ range.

Due to an increase in the overall amount of absorbing molecules, an increase in β causes the modulation characteristics to move towards lower transmission values [12]. The difference between two NTPI values (probe beam modulation) for two $I_m^{'}$ values initially grows and then falls with increase in β, and the decrease in the value of NTPI with β at a given $I_m^{'}$ is nonlinear. As a result, there is an ideal value of β for each of the two $I_m^{'}$ provided values at which the probe beam may be modulated to its peak value [12]. Figure 2 shows the nature of dependence of modulation of NTPI between pump intensities 0 and $I_m^{'}$, with β for different $I_m^{'}$ values. One may observe that optimum β decreases with rise in $I_m^{'}$ for decreasing NTPI part, and it increases with increase in $I_m^{'}$ for increasing NTPI part of modulation characteristics.

The analytical expression for optimum value of β (β_{opt}) for nonlinear absorbers is given in Eq. 1 and can be obtained for bR by substituting the value of $f_{bR}(I_m)$ from Eq. 2. The variation of β_{opt} (for maximum probe beam modulation, corresponding to the two pump intensity values 0 and $I_m^{'}$) with $I_m^{'}$ has been shown in Figure 3 on the left hand side scale. Figure 3 also shows the variation of the corresponding negative value of maximum modulation ($-\eta_{max}$) with $I_m^{'}$ on the right hand side scale. It is clear from Figures 1 and 3 that the

respective variation of NTPI, β_{opt} and $-\eta_{max}$ with $I_m^{'}$, follows the same trend. In case of NTPI possessing negative slope with $I_m^{'}$, β_{opt} and corresponding $-\eta_{max}$ decrease with rise in $I_m^{'}$ as $I_m^{'}$ compensates for increased absorption due to concentration or thickness. For positive slope of NTPI with $I_m^{'}$, β_{opt} and corresponding $-\eta_{max}$ increase with rise in $I_m^{'}$, as more concentration of absorbing molecules is required to stabilize the point of maximum contrast. For a range of $I_m^{'}$, parametric variation of β_{opt} and $-\eta_{max}$ with NTPI for $\beta = 0.9$ is shown in Figure 4 and is almost linear. Hence, the nature of the variation of β_{opt} and $-\eta_{max}$ with pump intensity, respectively, can be deduced by analysing the transmission characteristics of nonlinear absorbers.

In contrast to the pump beam, the probe beam's intensity is typically thought to be quite low in pump probe procedures. The probe intensity may have an impact on the modulation properties when there is significant overlap in the absorption spectra of intermediates. Strong probing beams can be used to change the nature of the modulation features because an absorption spectra of B and O-states exhibit substantial overlaps. We have also examined the modulation properties while taking into account the significant 640 nm probe beam transmission. For O-state dynamics, the probe intensity at 640 nm drives the photocycle and causes the rise in the population of O-state even when the pump beams. This leads to decrease in initial NTPI in non-appearance of the pump signal. The rise in $I_m^{'}$, here, results in decrease in population of O-state as it absorbs the optical signal at 570 nm and it finally leads to the monotonic enhancement in NTPI with $I_m^{'}$. Interestingly, in this case also, the variation of NTPI, β_{opt} and corresponding $-\eta_{max}$ with $I_m^{'}$ follows the same trend.

The kinetic and spectral parameters of bR can be modified via chemical, physical, and genetic engineering techniques [10-12]. The impact of different spectral and kinetic parameters has been examined on β_{opt} for modulation characteristics of bR having negative slope with $I_m^{'}$. The rise in absorption cross-section of initial B-state at the wavelength 570 nm, results in decrease in β_{opt} for given $I_m^{'}$ value. Increasing values of absorption cross-section of excited O-state at probe wavelength results in decrease in β_{opt}.

4. Conclusions

We have presented a general analytical expression for the optimum value of β (β_{opt}) for nonlinear absorbing thin samples. As a specific example, we consider all-optical light modulation in bR, that exhibits a complex photocycle [7, 8]. We have also analysed in detail the effect of the conversion between reverse saturable absorption (RSA) and saturable absorption (SA) in bR on β_{opt}. The effect of various spectral and kinetic parameters and pump and probe intensity on β_{opt} for maximum modulation (η_{max}) has also been studied. It is shown that β_{opt} decreases and its corresponding η_{max} increases with increase in pump intensity for reverse saturable absorbers at probe wave length, whereas β_{opt} increases for saturable absorbers. Interestingly, the respective variation of normalized transmitted probe intensity, β_{opt} and $-\eta_{max}$ with pump intensity, follows the same trend. Hence, the nature of the variation of β_{opt} and η_{max} with pump intensity, respectively, can be deduced by analysing the transmission characteristics of nonlinear absorbers. The analysis would be useful to design nonlinear absorption based all-optical devices.

Acknowledgements

The author is very thankful to the Eminent Referees for many useful suggestions to carry out this research work.

References

[1] A. Liu, R. Jones, L. Liao, D.S. Rubio, D. Rubin, O. Cohen, R. Nicolaescu, M. Paniccia, A high speed silicon optical modulator based on a metal-oxide semiconductor capacitor, *Nature* **427** (2004) 615-628.

[2] V.R. Almeida, C.A. Barrios, R.R. Panepucci, M. Lipson, All-optical control of light on a silicon chip, *Nature* **431** (2004) 1081-1084.

[3] J C. Li, J. Si, M. Yang, R. Wang, L. Zhang, Excited state nonlinear absorption in multi-energy-level molecular systems, *Phys. Rev. A* **51** (1995) 569.

[4] P. Sharma, S. Roy, C.P. Singh, Low power spatial light modulator with pharaonis phoborhodopsin, *Thin Solid Films* **477** (2005) 227-232.

[5] S. Roy, T. Kikukawa, P. Sharma, N. Kamo, All-optical switching in pharaonis phoborhodopsin protein molecules, *IEEE Trans. Nanobiosci.* **5** (2006) 178.

[6] P. Sharma, S. Roy, All-optical light modulation in pharaonis phoborhodopsin and its applications to parallel logic gates, *J. Appl. Phys.* **96** (2004) 1687.

[7] D. Dantsker, S. Speiser, Utilization of photoreversible optical nonlinearities intrans-cis photochromic molecules for spatial light modulation, *Appl. Phys. B* **58** (1994) 97-104.

[8] M. Brunel, K.A. Ameur, F. Sanchez, Optimization of the transverse concentration profile in optical limiting devices, *Opt. Commun.* **187** (2001) 271-276.

[9] T. Xia, D.J. Hagan, A. Dogariu, A.A. Said, E.W.V. Stryland, Optimization of optical limiting devices based on excited state absorption, *Appl. Opt.* **36** (1997) 4110-4122.

[10] N. Hampp, Bacteriorhodopsin as a photochromatic retinal protein for optical memories, *Chem. Rev.* **100** (2000) 1755.

[11] S. Roy, C.P. Singh, K.P.J. Reddy, Generalized model for all-optical light modulation in bacteriorhodopsin, *J. Appl. Phys.* **90** (2001) 3679.

[12] P. Sharma, S. Roy, All-optical biomolecular parallel logic gates with bacteriorhodopsin, *IEEE Trans. Nanobiosci.* **3** (2004) 129.

[13] J. A. Stuart, D.L. Mercy, K.J. Wise, R.R. Birge, Volumetric optical memory based on bacteriorhodopsin, *Synth. Metals* **127** (2002) 3-15.