Eliska Jurisova – Jarmila Mullerova *

NUMERICAL STUDIES OF OPTICAL BISTABILITY OF ABSORPTIVE TYPE

Theoretical analysis is presented to search for the purely absorptive optical bistability in a Fabry-Perot cavity filled with a saturable absorber. A simple model of singlet absorption is used on numerically solving the relationships between the input and output light fields. The proposed model corresponds well to organic compounds considered as novel optical materials with third-order optical nonlinearities. The effects of saturable absorber parameters on the output light characteristics are studied and purely absorptive optical bistabilities are shown numerically to search for bistable behaviour. The simulated results are indications for promising applications in optical switches for all-optical communication networks.

1. Introduction

Optical bistable devices have a large number of applications in all-optical communication systems. They are building blocks of optical logic, memories, switches, light power limiters etc. that are crucial parts of optical network nodes [1–3]. Optical bistability is a phenomenon in which the bistable device exhibits two stable output states under one input state. The optical bistability arises from the energy exchange between nonlinear optical medium and electromagnetic field. The system is said to be bistable when it can assume two output stable states for the same value of an applied input signal over some range of input signal values, Optical bistability requires a nonlinear medium in presence of feedback. When the nonlinearities are optically-induced, and the feedback is also provided in the optical domain, an all-optical device can be fabricated [4]. The phenomenon of absorptive optical bistability was first theoretically predicted in 1969 by Szoke [5]. Third-order nonlinear materials are mainly metal particles, semiconductors including those of quantum confinement and organics [6–8]. Among organic materials, π-conjugated compounds attract attention due to unique electrical and optical properties including large third-order optical nonlinearities, low-cost manufacturing and flexibility of properties modifications by chemical synthesis as well as deposition techniques. There has been a prolonged research of all-optical switching in organic devices based on optical bistabilities [9]. Parameters of nonlinear medium should be properly chosen to achieve the optical bistability. For the construction of optical bistable devices of absorptive type, saturable absorbers and positive feedback are necessary. A lot of studies report on nonlinear absorption in a great variety of organic compounds [10, 11]. Therefore, the prediction of their bistable behaviour based on theoretical calculations with real experimental data gathered on specific organics is of interest for designing optically bistable devices.

This contribution presents numerical studies of purely absorptive optical bistabilities in saturable absorbers in the Fabry-Perot cavity. Organic dyes with spectral sensitivities such as xanthene, polymethene or coumarin dyes are known for their nonlinear optical properties including saturable absorption. In this paper, we predict and numerically demonstrate the optical bistability in two organic dyes of the xanthenes type dispersed in glass and polymethene dyes dissolved in organic solvents. We simulated their bistable behaviour considering absorption parameters and positive feedback conditions with experimental data from [8, 11].

2. Theory

A simple purely absorptive bistable device can be constructed by placing nonlinearly absorbing medium inside a resonator, e.g. Fabry-Perot cavity (Fig.1).

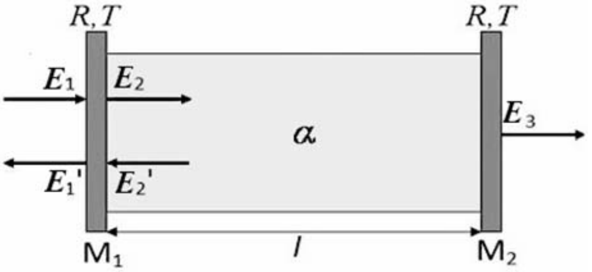


Fig. 1 Fabry-Perot cavity filled with nonlinear absorber of the nonlinear absorption coefficient α and the thickness l. M_1, M_2 are mirrors of reflectance R and transmittance T. The notifications of electric fields are obvious from the scheme

---

* **Eliska Jurisova, Jarmila Mullerova**
Department of Engineering Fundamentals, Faculty of Electrical Engineering, University of Zilina, Liptovsky Mikulas, Slovakia,
E-mail: jurisova@lm.uniza.sk

The cavity is formed by two parallel identical flat highly reflective mirrors and demonstrated to provide optical bistability.

The absorption coefficient α is an important wavelength-dependent optical parameter expressing the decrease dI of the light intensity I in the length dz of the absorbing medium

$$dI = -\alpha I dz \tag{1}$$

Eq. (1) is the well-known differential form of the Lambert-Beer law. It is easy to integrate if α is intensity-independent. However, in a nonlinear absorber, α depends on the light intensity. If the intensity of the input radiation is intense enough, the absorption of the medium decreases the consequence of what is the increase of the medium transmittance up to the saturation. If the saturation does not occur and at high intensities decreases again, the so-called reverse saturable absorption occurs. This effect can be used as optical power limiting [12]. According to the absorptive optical bistability model of Szoke [5], the amplitude of electric field entering the Fabry-Perot cavity at normal incidence, electric fields inside the cavity and the output field (Fig.1) obey the following equations

$$E_2 = \tau E_1 + \rho E_2' \tag{2}$$

$$E_1' = \rho E_1 + \tau E_2' \tag{3}$$

$$E_2' = \rho E_2 e^{-2ikl} e^{-\alpha l} \tag{4}$$

$$E_3 = \tau E_2 e^{-2ikl} e^{\frac{-\alpha l}{2}} \tag{5}$$

where l is the absorber thickness, k is the wavenumber, ρ and τ are Fresnel amplitudes of mirror reflectance and transmittance. If $e^{-2ikl} = 1$ (the Fabry-Perot resonance condition), the relationship between the electric field inside the Fabry-Perot cavity and the input electric field can be expressed as [5, 13]

$$E_2 = \frac{\tau E_1}{1 - \rho^2 e^{-\alpha l}} \tag{6}$$

Then for the light intensities ($I \sim E_2$) the following is valid

$$I_2 = \frac{T I_1}{\left(1 - \mathrm{Re}^{-l}\right)^2} \tag{7}$$

where mirror transmittance $T = \tau^2$, and reflectance $R = \rho^2$ ($R + T = 1$, $R >> T$). Moreover, if $\alpha l << 1$, the equation (7) can be expresses as

$$I_2 = \frac{I_1}{T\left(1 + \frac{\alpha l R}{T}\right)^2} \tag{8}$$

If there is a nonlinear absorber in the Fabry-Perot cavity, $\alpha = \alpha(I_2)$. From Eq. (5) we can see that in case of $\alpha l << 1$ the output electric field $E_3 = \tau E_2$. Then the output intensity is simply $I_3 = TI_2$. Therefore it is sufficient to analyze Eq. (8) to judge if the absorber of the specific absorptive nonlinearity $\alpha = \alpha(I_2)$ will exhibit the bistable output intensity I_3 as a function of I_1.

The simulations of Eq. (8) were performed with real experimental data for nonlinear organic absorbers and the prospects to achieve bistable performance were modelled. The constant spatial profile of the propagating pulses was supposed in our calculations and the optical pulse durations were considered to be larger than the lifetimes of the excited levels. Under this assumption the saturable absorbers act as fast absorbers and the fast relaxation approximation enables to expect their steady state behaviour.

3. Numerical Simulation

We numerically investigated the optical bistability in organic dyes of molecules that contain conjugated bonds with delocalized π – electrons that are responsible for large third-order nonlinear susceptibilities [9]:

- The xanthene dye of fluorescein dispersed in boric acid glass film (BAG) (with 10^{-4} M concentration) of optical parameters as in [11]. Fluorescein is a bright green fluorophore commonly used in fluorescence measurements. The maximum of the absorption spectrum is at ~ 460 nm.
- The xanthene dye of rhodamine 6G dispersed in boric acid glass film (BAG) (with 10^{-4} M concentration) of optical parameters taken from [11]. Rhodamine 6G is chromophore used often as a tracer or a laser dye. The maximum of the absorption spectrum is at ~ 515 nm.
- Polymethine dyes of cationic type dissolved in ethanol or polyurethane. Polymethine dyes are known as organic colorants and media laser technique. The maximum of the absorption spectrum is at ~ 1100 nm. The optical parameters for this study were taken from [8].

All investigated organic dyes dispersed in BAG or dissolved in organic solvents have energy level structure of singlet and triplet states typical for organic media (Fig. 2) with the singlet S_0 as the ground state.

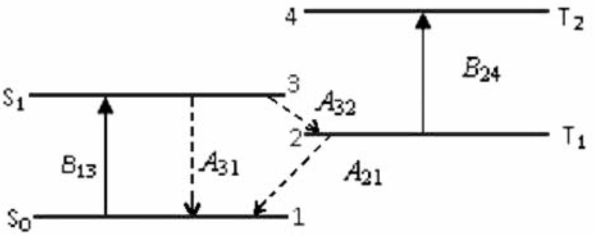


Fig. 2 Energy level model of singlets (S) and triplets (T) typical for organic dyes under study. The model shows optical excitations (upward-pointing arrows) of probabilities given by Einstein's coefficients B_{13}, B_{24}. A_{31}, A_{32}, A_{21} are Einstein's coefficients for the relaxations from excited states (downward-pointing arrows)

After quick excitation to S_1 the molecule may relax to S_0 with the probability $A_{21} = 1/\tau_{21}$ or to T_1 with the probability $A_{32} = 1/\tau_{32}$ (τ_{21}, τ_{32} are the relaxation times). The transition from T_1 to S_1 can be neglected. In liquid state, the dye molecules decay very quickly to from T_1 to S_0. On the contrary, in dye-doped solids, the $T_1 \rightarrow S_0$ decay is on the millisecond scale. Moreover, by adjusting the excitation wavelength only to the transition $S_0 \rightarrow S_1$, the triplet-triplet absorption $T_1 \rightarrow T_2$ does not occur.

With these assumptions, the absorption coefficient depends on the light intensity I2 in the cavity as follows

$$\alpha(I_2) = \frac{\alpha_0}{1 + \beta I_2} \tag{9}$$

In Eq. (9) α_0 is the absorption coefficient at low intensities of light, β is the so-called parameter of nonlinearity. For the energy level model in Fig. 2 the parameter β is [11]

$$\beta = \frac{\sigma_{13}}{h\nu_{13}} \frac{\tau_{21}\tau_{31}}{\tau_{32} + \tau_{31}} \tag{10}$$

where $\tau_{31} = 1/A_{31}$, σ_{13} is the absorption cross-section for $S_0 \rightarrow S_1$, h is the Planck's constant and ν_{13} is the frequency corresponding to the $S_0 \rightarrow S_1$ excitation. After substituting Eq. (9) into Eq. (8), we have the following transcendental relationship between I_2 and I_1

$$I_2 = \frac{I_1}{T\left(1 + \dfrac{\alpha_0 l R}{T(1 + \beta I_2)}\right)^2} \tag{11}$$

The product $\alpha_0 l$ of the linear absorption coefficient α_0 and the medium length l is the so-called linear absorbance that is related to the initial linear transmittance T_0 of the sample as $T_0 = \exp(-\alpha_0 l)$.

The values of $\alpha_0 l$ and the parameter of nonlinearity β for ten samples of fluorescein, four samples of rhodamine 6G doped BAG [11] and for three solutions of polymethine dye structures, namely cationic [7] used for the calculations are in Table 1.

In the first course of simulations the absorbers were settled in the Fabry-Perot cavity of the mirror reflectance of $R = 0.9$ and the mirror transmittance $T = 1 - R$.

Table 1: Optical absorption parameters for fluorescein, rhodamin 6G doped BAG films [11] and for solutions of polymethine dye solutions [8]

	BAG doped by Fluorescein		BAG doped by Rhodamine 6G		Solutions of polymethine dyes	
	$\alpha_0 l$	β (cm^2/W)	$\alpha_0 l$	β (cm^2/W)	$\alpha_0 l$	β (cm^2/W)
1	0.247	62.50	0.79	0.463	2.905	$2.50 . 10^{-9}$
2	0.534	18.52	0.95	0.452	2.705	$2.94 . 10^{-7}$
3	0.699	38.46	1.79	0.418	1.520	$2.94 . 10^{-7}$
4	0.814	26.32	3.47	0.420		
5	1.070	55.56				
6	1.390	37.04				
7	1.680	40.00				
8	2.030	52.63				
9	2.310	38.46				
10	2.680	43.48				

4. Results and Discussion

The numerical study of the relationship between I_1 and I_2 according to Eq. (11) was performed under typical input powers from [10]. Results show that the optical bistability depends not only on nonlinear parameter β but also on linear absorbance α0l (Fig. 3). For the values of $\alpha_0 l > 1.39$ for fluorescein the output intensity I_2 is a two-valued function of I_1 and the hysteresis loops increase. The simulation shows that fluorescein under these conditions would exhibit bistable behaviour even with relatively low input intensities of light. From Fig. 3a) for fluorescein dispersed in BAG we also see that onset of the optical bistability for increasing $\alpha_0 l$ requires greater incident light intensities I_1.

The similar results are in Fig. 3b) for rhodamine 6G. The optical bistability could be achieved at input intensities > 20 W/cm^2 at $\alpha_0 l > 1.79$. The input intensities necessary for the optical bistability are higher than those for fluorescein which could be explained by lower values of parameter β representing the nonlinearity of rhodamine 6G in comparison with fluorescein.

The optical bistability is evidenced in solutions of polymethine dyes shown in Fig. 3c) and 3d). However, the input intensities necessary for the bistable performance are much higher in comparison with fluorescein and rhodamine 6G because the nonlinearity represented by the parameter β is very small (Table 1).

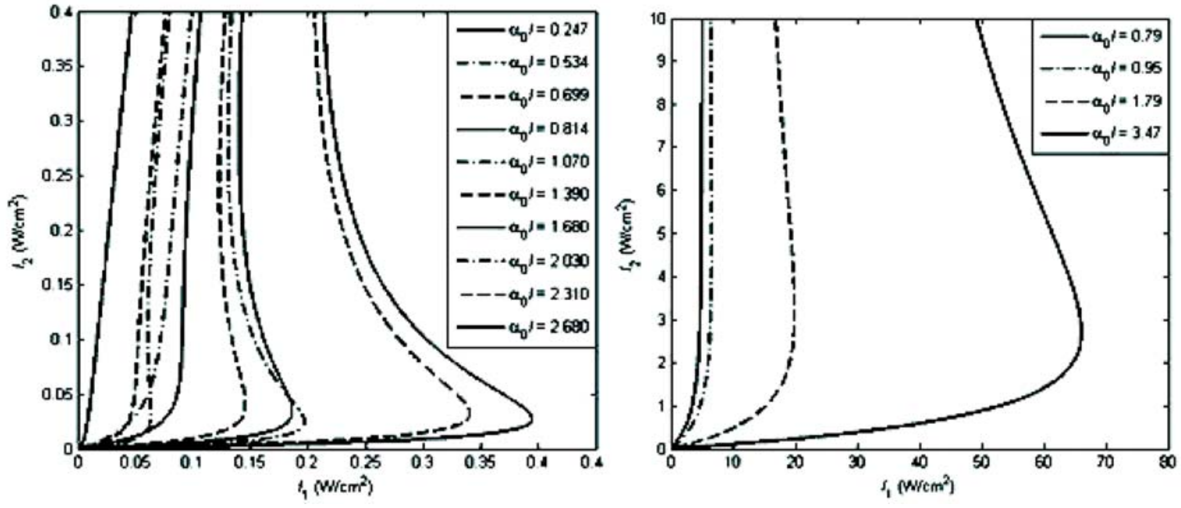
In the next course of simulations we investigated the influence of the Fabry-Perot mirror reflectance on the bistable behaviour. Several different values of the mirror reflectance from the range of 0.6 ÷ 0.98 were used (Figs. 4 and 5).

From Fig. 4a we can see that fluorescein requires mirrors reflectance at least 0.85 to exhibit slight bistability in the Fabry-Perot cavity. In Fig. 4b the numerical analysis for rhodamine 6G with the same mirror reflectance is shown. We found out that a potential bistable device based on rhodamine 6G of the above $\alpha_0 l$ should be placed in the resonator of the mirror reflectance $R = 0.9$ at least. To improve the data visualisation, the numerical results in 3D are illustrated in Fig. 4 as well. The results proved the important role of the positive feedback in an absorptive bistable device.

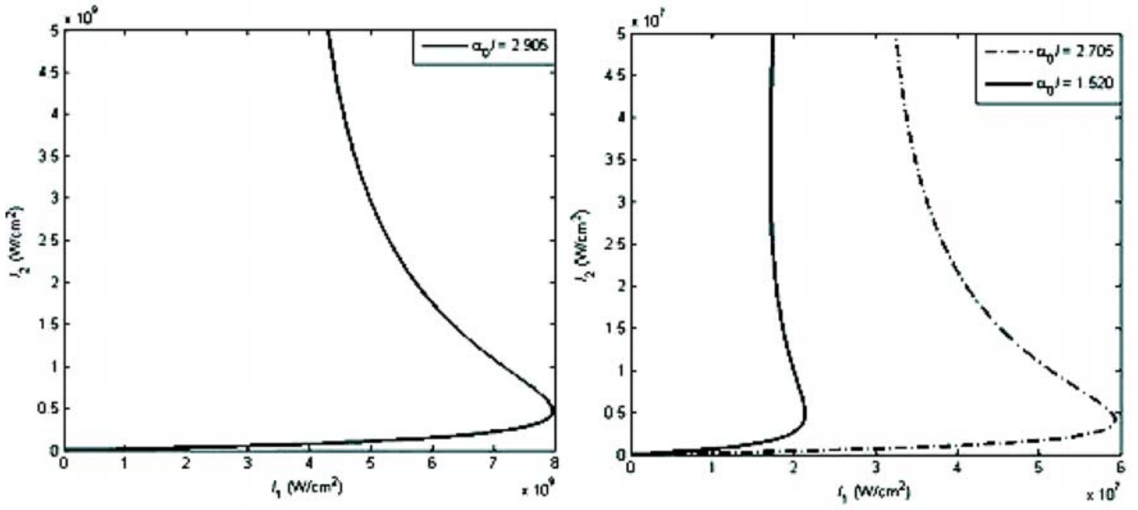
The bistable behaviour was numerically investigated in solutions of polymethine dyes (Fig. 5). We see that increasing reflectance R facilitates the optical bistability occurrences, although at very high input light intensities.

The absorption lines of organic dyes are often inhomogeneously broadened inside a host material [6].The inhomogeneous broadening of optical transitions originates from the dye – matrix interaction, especially from different positions of molecules in the matrix. This could occur in case of dye molecules dispersion in glass matrix. On that occasion the intensity-dependent absorption coefficient is given by the following modification of Eq. (9)

$$\alpha(I_2) = \frac{\alpha_0}{\sqrt{1 + \beta I_2}} \tag{12}$$



a) fluorescein *b) rhodamine 6G*



c) polymethine dye solution 1 *d) polymethine dye solutions 2 and 3*

Fig. 3 Investigation of the optical bistability in a) fluorescein, b) rhodamine doped BAG and c) polymethine dye solution 1 and d) solutions of polymethine dyes 2 and 3; R = 0.9

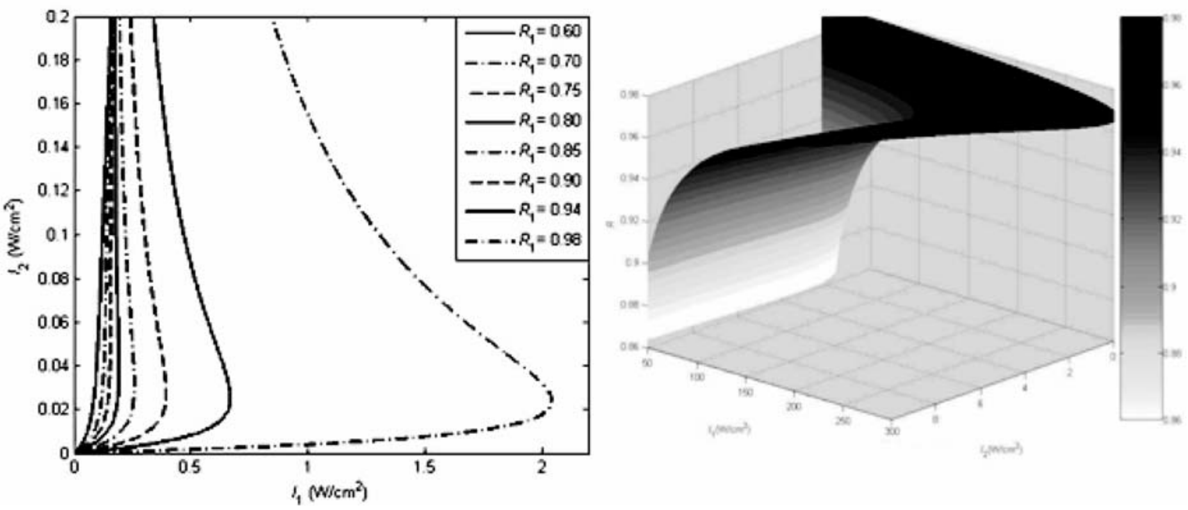
Due to inhomogeneous absorption line broadening expressed by Eq. (12) the following relationship between I_2 and I_1 as a result of substituting Eq. (12) into Eq. (8) is valid

$$I_1 = TI_2\left(1+\frac{\dfrac{\alpha_0 lR}{T}}{\sqrt{1+\beta I_2}}\right)^2 \qquad (13)$$

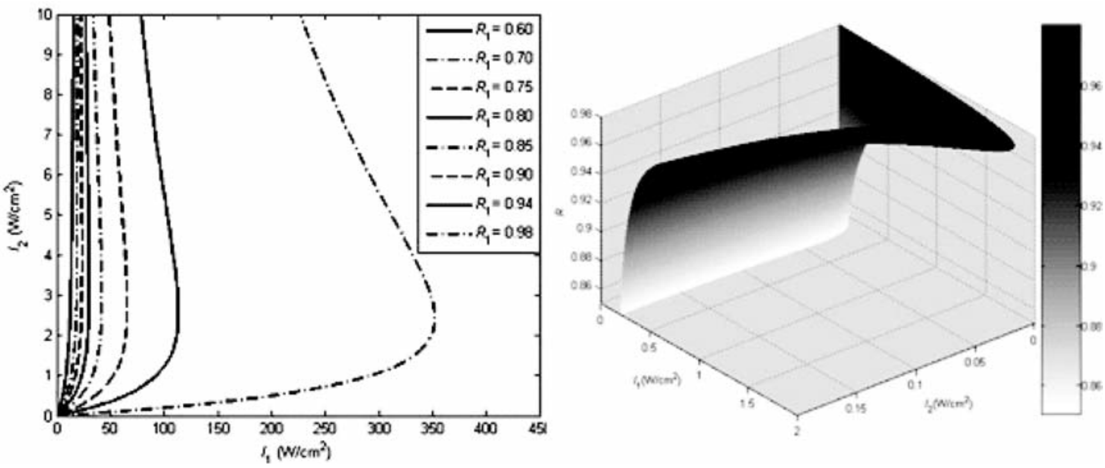
The results of numerical studies of the relationships between I_1 and I_2 according to Eq. (13) are in Fig. 6 for BAG doped by fluorescein and for solutions of polymethine dyes. We found out that the inhomogeneous broadening of the optical transitions results in the decay of possible bistable behaviour. Similar results were obtained for rhodamine 6G. Then either the inhomogeneous broadening should remain small or a thorough choice of parameters of the nonlinear medium with inhomogeneous broadening is necessary to achieve the bistable performance. This conclusion is in agreement with the calculations in [15].

4. Conclusion

We numerically analyzed the optical bistable behaviour of organic dyes. It is clearly shown that the bistable behaviour is influenced by the linear absorbance, the parameter of medium nonlinearity and by the Fabry-Perot mirror reflectance as a parameter of positive feedback of a optical bistable device. We numerically established linear absorbance thresholds for the optical bistability in fluorescein and rhodamine 6G doped glass and for polymethine dyes dissolved in organic solvents. At specific values of linear absorbance of all organic dyes, we numerically determined the

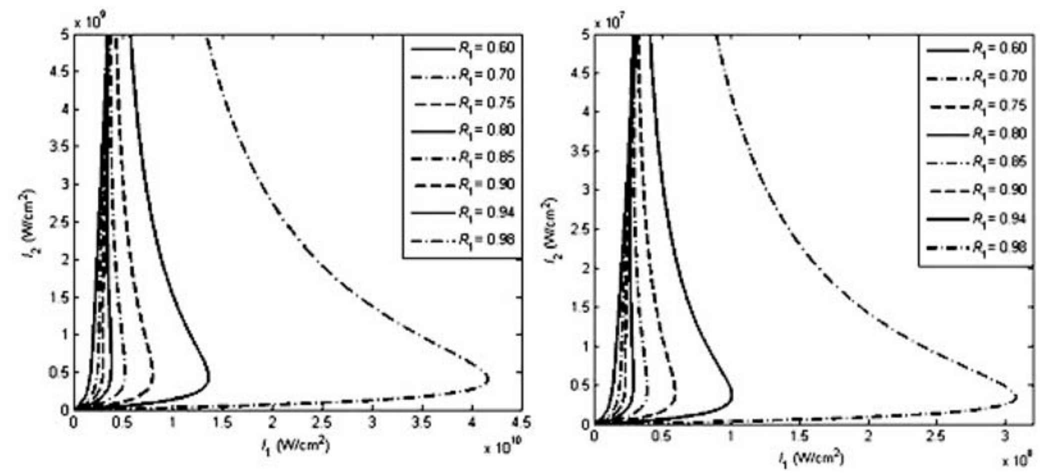


a) fluorescein at $\alpha_0 l = 2.68$



b) rhodamine 6G at $\alpha_0 l = 3.47$

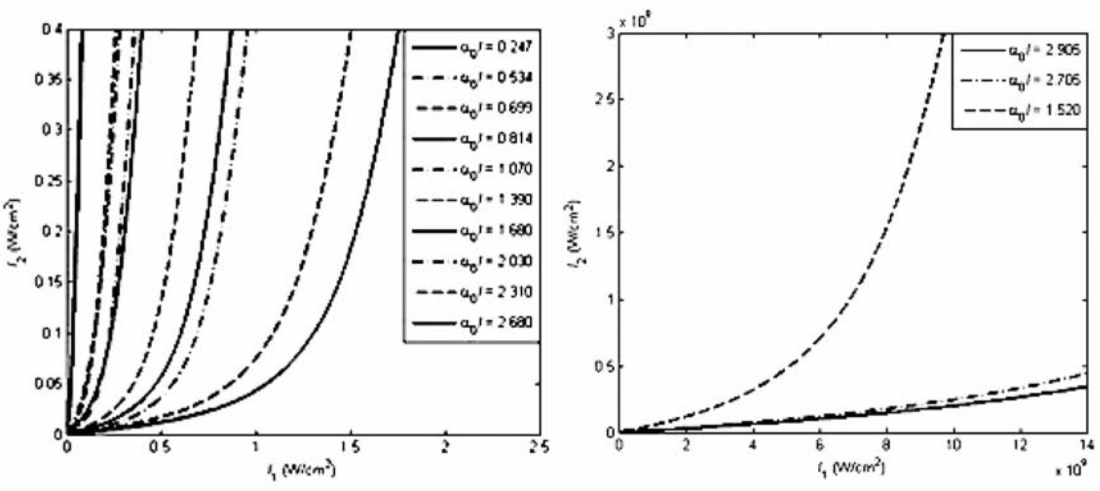
Fig. 4 Results 2D and 3D simulating optical bistability in BAG doped organic dyes



a) polymethine dye 1 at $\alpha 0_l = 2.905$

b) polymethine dye 2 at $\alpha_0 l = 2.705$

Fig. 5 Results simulating optical bistability in solutions of polymethine dyes



a) fluorescein

b) polymethine dye solutions

Fig. 6 Investigation of the optical bistability with inhomogeneously broadened spectral lines (varied $\alpha0l$ and β, $R = 0.9$)

optical bistability threshold of the cavity mirror reflectance. Additionally we found out that in case of inhomogeneous broadening of spectral lines related to the optical excitations and relaxations, the optical bistability is quenched in all investigated organic dyes.

Acknowledgement

This work was partly supported by Slovak Grant Agency under the project No. 1/0411/10.

References

[1] DADO, M., KRAJCI, S., DUBOVAN, J., SABOL, D.: Numerical Investigation of Optical Burst Switching, *Communication - Scientific letters of the University of Zilina,* pp. 20-24, No. 2, 2008.

[2] HENKER, R. et al.: A Review of Slow- and Fast- Light Based on Stimulated Brillouin Scattering in Future Optical Communication Networks. *Communication - Scientific letters of the University of Zilina*, pp. 45-52, No. 4, 2008.

[3] LIU, JIA-MING: *Photonic Devices.* Cambridge University Press, First published 2005.

[4] BHARGAVA, S. et al.: *Optical Bistability in a Nonlinear Resonator With Saturable Losses and Intensity- Dependent Refractive index.* Int'l Conf. on Computers and Devices for Communication, 2009.

[5] SZOKE, I. A., DANEU, V., GOLDHAR, J., KURNIT, N. B.: Bistable Optical Element and its Applications. *Appl. Phys*. Lett. 15, pp. 376-379, 1969.

[6] NIE, WENJIANG: Optical Nonlinearity: Phenomena, Applications, and Materials. *Advanced Materials* 5 (7/8), pp. 520-545, 1993.

[7] MATTU, J., JOHANSSON, T., LEACH, G.W.: Third Order Nonlinear Optical Response from Polythiophene-based thin Films. *J. Phys. Chem.*, 111, pp. 6868-6874, 2007.

[8] GANEEV, R.A. et al.: Characterization of Nonlinear Optical Parameters of Polymethine Dyes. *Appl. Phys.* B 76, pp. 683-686, 2003.

[9] XUE, J., FORREST, S.R.: Organic Optical Bistable Switch. *Appl. Phys.* Lett. 82, pp. 136-138, 2003.

[10] UNNIKRISHNAN, K.P., THOMAS, J., NAMPOORI, V.P.N., VALLABHAN, C.P.G.: Nonlinear Absorption in Certain Metal Phtalocyanines at Resonant and Near Resonant Wavelength. *Optics Communications* 217, pp. 269-274, 2003.

[11] SHARAN, A., SHARMAN, R. C., SANDHYA, S. N., AYYER, A., SHARMA, K. K.: Modeling Absorption in Saturable Absorbers. *Optics Communications* 199, pp. 267-275, 2001.

[12] HU, A.T. et al.: Reverse Saturable Absorption of Copper Phthalocyanines in Toluene and Solgel Tetraethyl Orthosilicate/polyvinyl Butyral Hybrid Film. *Dyes and Pigments* 62 (1), pp. 11-19, 2004.

[13] DJABI, S., DJABI, M.: Study of Optical Bistability in a Laser Containing a Saturable Absorber. *J. of Engineering and Applied Sciences* 2 (9), pp. 1383-1386, 2007.

[14] RENGE I., WILD, U. P.: Inhomogeneous Broadening and Pressure Shifts of the Optical Spectra in Organic Glasses at Low Temperatures. *J. Luminescence* 86 (3-4), pp. 321-247, 2000.

[15] BENIACHE, A., MOUASSA, S., VAN HOANG, D.: Optical Bistability of Fabry-Perot LSA with Inhomogeneous Broadening. *J. Engineering and Appl. Sciences* 2 (1), pp. 66-72, 2007.