J. At. Mol. Sci.
doi: 10.4208/jams.032714.062614a

Vol. 5, No. 3, pp. 254-262
August 2014

Optical limiting and dynamical two-photon absorption for femtosecond laser pulses in BDBAS

Yu-Jin Zhang, Yong Zhou*, Jun-Yang Ma, Yao Chen, and Chuan-Kui Wang

College of Physics and Electronics, Shandong Normal University, Jinan 250014, China

Received 27 March 2014; Accepted (in revised version) 26 June 2014
Published Online 8 August 2014

Abstract. By numerically solving the Maxwell-Bloch equations using an iterative predictor-corrector finite-difference time-domain (FDTD) technique, the optical limiting (OL) and two-photon absorption (TPA) properties for femtosecond laser pulses in a strong TPA organic molecular (4,4'-bis (di-*n*-butylamino) stilbene (BDBAS)) medium are studied. The medium has obvious OL behavior in a certain intensity region which is dependent on the propagation distance, sample concentration, input pulse width and effect of optical ionization. Moreover, dynamical TPA cross sections are obtained, showing an increasing trend with the broadening of the pulse width and the propagation distance in the femtosecond time domain.

PACS: 33.80.-b, 82.50.Pt, 31.15.A-
Key words: optical limiting, two-photon absorption, organic molecule

1 Introduction

The interaction between light and matter has always been evoking one's curiosity and motivation [1-4]. On the basis of the advances in ultra-short and ultra-intense laser technology, it is available to generate extremely short and intense pulses [5,6], which pushes the nonlinear optics into a new stage [7-9]. In recent years, studies of OL have been more and more interesting due to the need for protection of the optical sensors against intense laser radiation and for peak-power stabilization of the intense laser pulses [10-12]. One of the key challenges in the development of optical limiters is to search for appropriate materials that rapidly respond to the applied laser intensity, own good nonlinear absorption characteristics over a broad range of wavelengths, and have a high damage threshold. With the progress of design and synthesis technology of organic materials, organic

*Corresponding author. *Email address:* zhangyujin312@163.com (Y. Zhou)

conjugated compounds have shown great promise for OL applications [10,13-15]. They possess various nonlinear mechanisms to produce OL, in which TPA is the major one.

As a typical organic conjugated compound, BDBAS were demonstrated to have obvious nonlinear optical properties [16]. In this work, taking BDBAS as the sample, we numerically study the dynamical behavior of OL caused by TPA and determine the dynamical TPA cross section when ultra-short laser pulses are used. Also, the influences of propagation distance, sample concentration, effect of optical ionization and pulse width on OL are discussed.

2 Theoretical methods

2.1 Maxwell-Bloch equations for a three-level system

Based on the semi-classical theory on the interaction between laser pulses and molecules, the electromagnetic radiation can be described classically by Maxwell equations, and the molecular system can be treated by Bloch equations quantum mechanically.

We assume that the incident electromagnetic field is polarized along the x-axis and propagates along the z-axis to an input interface of the medium at z=0. Then the Maxwell equations can take the form as follows:

$$\begin{aligned}\frac{\partial E_x}{\partial z} &= -\mu_0 \frac{\partial H_y}{\partial t}\\ \frac{\partial H_y}{\partial z} &= -\frac{\partial P_x}{\partial t} - \varepsilon_0 \frac{\partial E_x}{\partial t}\end{aligned} \tag{1}$$

where μ_0 and ε_0 are the permeability and permittivity of free space, respectively.

Taken the relaxation effect and ionization effect into consideration, the density matrix equation can be written as:

$$\begin{aligned}\dot{\rho}_{mn} &= -\frac{i}{\hbar}[\hat{H},\hat{\rho}]_{mn} - \gamma_{mn}\rho_{mn} - \frac{\gamma_{ph}^{(m)}(t)+\gamma_{ph}^{(n)}(t)}{2}\rho_{mn} \qquad (m \neq n)\\ \dot{\rho}_{nn} &= -\frac{i}{\hbar}[\hat{H},\hat{\rho}]_{nn} + \sum_{E_m>E_n} \Gamma_{nm}\rho_{mm} - \sum_{E_m<E_n} \Gamma_{mn}\rho_{nn} - \gamma_{ph}^{(n)}(t)\rho_{nn}\end{aligned} \tag{2}$$

here Γ_{nm} gives decay rate of the population from level m to level n, and γ_{mn} is the relaxation rate of the density matrix element ρ_{mn}. $\gamma_{ph}^{(m)}(t)$ and $\gamma_{ph}^{(n)}(t)$ are the ionization rate of level m and level n, respectively. It can be expressed as $\gamma_{ph}^{(i)}(t) = \sigma_i I(t)/(\hbar\omega_p)$, where $I(t) = c\varepsilon_0 \mid E(t,z) \mid^2/2$ is the laser intensity, σ_i is the ionization cross section of level i, which has an order about 10^{-20} m^2 [17], and ω_p is the frequency of the input pulse. $\hat{H}$ is the Hamiltonian of the system, which can be expressed as the sum of free Hamiltonian and interaction Hamiltonian. Within the dipole approximation, it can be written as:

$$\hat{H} = \hat{H}_0 + \hat{H}' = \hat{H}_0 - \hat{\mu}\cdot E. \tag{3}$$