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Modulation on optical limiting and two-photon absorption behavior of a molecular medium by two-color ultrashort laser pulses

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Abstract. The dynamical behavior of two-color ultrashort laser pulse propagating in a molecular medium 4,4'-bis(di-*n* -butylamino)stilbene (BDBAS) has been studied by employing an iterative predictor-corrector finite-difference time domain (FDTD) technique to solve the Maxwell-Bloch equations. The results indicate that the spectrum of electric field shows oscillation feature around the two-photon resonant frequency when the delay time exists due to the interaction between two sub-pulses. The optical limiting window turns narrower and the saturation value of output intensity gets larger with the presence of delay time compared with no delay time case. The dynamical TPA cross section is smaller with the increase of delay time. The studies suggest a method to modulate the nonlinear optical properties of the medium by controlling the delay time of the two-color pulse.

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Key words: optical limiting, two-photon absorption, Maxwell-Bloch equations, two-color pulse

1 Introduction

During the past few decades, advances in ultrafast laser technology have made the generation of extremely short and intense pulses with only few optical periods in the visible region to be possible[1]. At the same time, studies on the intense fields-nonlinear materials interaction[2-4] and new mechanisms for generating shorter pulses were demonstrated theoretically[5]. High power laser sources have motivated an extensive research for the design of optical limiting (OL) system in order to protect sensors and human eye. There are several different mechanisms lead to OL behavior, such as reverse saturable

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absorption (RSA)[6], nonlinear refraction, optically induced scattering[7], and most importantly, two-photon absorption (TPA)[8]. Based on the *ab initio* level, several methods have been developed to calculate the intrinsic TPA cross section[9-10]. However, the actual TPA cross section of the medium is measured in laser field, which means its value is strongly dependent on the dynamical parameters of the pulse, such as pulse width, pulse intensity[8,11]. In recent years, lots of attentions are paid on developing approaches for modulating the OL behavior and TPA cross section.

Currently, there is a growing interest to investigate coherent control of nonlinear optical process with two strong laser fields, leading to numbers of interesting phenomena, such as controlling ionization, spontaneous radiation, population inversion, high-order harmonic generation (HHG) and so on. He *et al.* studied the HHG in argon using 800 nm and 400 nm laser fields simultaneously. They found that the interference of the two-color field could strongly modulate the intensity and divergence of the emitted even and odd harmonics, which is a function of the relative delay between the two fields[12]. Wu *et al.* demonstrated that the THz generation from a two-color pulse composed of the fundamental and second-harmonic waves can be coherently controlled by field-free molecular alignment[13]. Song *et al.* theoretically investigated the two-color interference effects for ultrashort laser pulses propagating in a two-level medium and the formation of higher spectral components[14]. The studies above motivate us to modulate the OL and TPA of the organic molecular materials using two-color pulses. The overlap and interference effects between the two pulses would lead to mutually interaction while propagating, and the relative phase and delay time between the two laser pulses can be used to modulate the nonlinear process.

In this paper, we study the dynamical behavior of two-color ultrashort laser pulse in a cascade three-level molecular BDBAS medium by solving the Maxwell-Bloch equations beyond the slowly-varying envelope approximation and the rotating-wave approximation. The spatial evolution properties of the pulse are analyzed, and the OL as well as dynamical TPA cross sections of the medium are given.

2 Theoretical methods

2.1 Maxwell-Bloch equations for a three-level system

The theoretical details are referred to in Ref. [15-16]. Here, we just list the main formulations. We treat the electromagnetic classically, and the molecule quantum mechanically. Then the electromagnetic radiation can be described by Maxwell equations, and the molecular system can be depicted by Bloch equations.

Starting with the density matrix equation with relaxation effect:

$$\dot{\rho}_{mn} = -\frac{i}{\hbar}[\hat{H},\rho]_{mn} - \gamma_{mn}(\rho_{mn} - \rho^0_{mn}) \tag{1}$$

where γ_{mn} is the relaxation rate of the density matrix element ρ_{mn}. $\hat{H}$ is the Hamilton of