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Nonlinear absorption study in four and five energy level systems

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ABSTRACT

Nonlinear absorption in four and five energy level systems have been studied with the aid of steady-state rate equation approach. We report on the tunability of saturable and reverse saturable absorption as a function of spectroscopic parameters (lifetimes and absorption cross-sections). Detailed information is given on the estimation of spectroscopic parameters in the nonlinear absorption spectroscopy. The advantage of four and five level cascade models over other models is that one can produce simultaneously saturable and reverse saturable absorption. Four and five level models can be potentially used as optical limiters and saturable absorbers due to their merit over two and three level models in terms of long tunability and high power control.

1. Introduction

In past two decades, nonlinear absorption attracted a great interest in the optical field due to its wide range of applications in the spectroscopic study. The spectroscopic parameters (lifetimes and absorption cross-sections) can be measured through pump-probe absorption spectroscopy [1,2], single beam transmittance (SBT) [3–5] and z-scan [6–8]. While, pump-probe deals with non-degenerate nonlinear absorption spectroscopy, z-scan and SBT are used in degenerate nonlinear absorption spectroscopy. Among these techniques, z-scan is a very simplest one in the spectroscopic study. However, this technique is insufficient, when it comes to the more than three energy levels contributed in the absorption process due to its finite longitudinal intensity profile restricted by focusing lens. In the z-scan, the sample has to cross the focal plane in the scanning process and the intensity at focus can damage the sample. To avoid the sample damage, while it is crossing the focus, we have to focus very low power and which is insufficient to study the higher excited states. But through SBT one can overcome this flaw by its extended tunable range which is mainly determined by laser power. The requirement of high power in SBT can be achieved by present available high power lasers without focusing.

Experimentally well studied the four and five energy levels participation in the degenerate absorption process [9–20]. But still, no article has discussed absorption cross-sections and lifetimes effect in the nonlinear absorption process. In this article, through rate equation approach, we have shown, how one can see the single saturable absorption (SSA) [21], double saturable absorption (DSA) [21] and saturable with reverse saturable absorption (SRSA). In addition to this, systematically studied the influence of spectroscopic parameters in the transmittance curve through SBT. Besides the rate equation method intrinsic beauty in the understanding of energy levels population dynamics in the materials in the presence of coherent optical beams excitation, it is an accomplished method in the understanding of nonlinear absorption spectroscopy. Rate equation technique is the efficient and simplest one to use to study the material absorption in case of four and five levels participation in the absorption process. Throughout this article, following parameters are considered for simulation: excitation wavelength (λ) is 532 nm, interacting length of the material with the laser beam (L) is 0.1 mm and number density of absorption species (N) is 10^{19} absorption species/cm^3. The number density of molecules in each state $|i\rangle$ is taken to be N_i with satisfying the condition $N = \Sigma N_i$ and the fractional number density in each state is $n_i = N_i/N$ and $\Sigma n_i = 1$.

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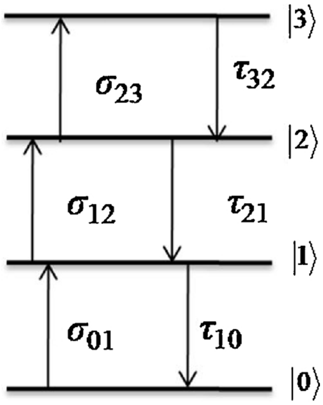

Fig. 1. Four level cascade system.

The pumping rate from $|i\rangle$ to $|j\rangle$ level is $W_{ij}=(I/h\nu)\sigma_{ij}$, where σ_{ij} is the absorption cross-section from $|i\rangle$ to $|j\rangle$ level. The decay time from $|j\rangle$ to $|i\rangle$ level is τ_{ji}. In all the models, the rate equations has been solved by considering steady state population i.e., $dn_i/dt = 0$. The transmittance of the interacting material is given by $T = \exp(-\alpha_{eff}L)$, where α_{eff} is effective absorption coefficient, which is differer for each energy model.

2. Four level cascade system

In four level cascade systems, subsequent energy levels are separated by equal energies (E_j-E_i = photon energy, $j=i+1$) as depicted in Fig. 1. Single frequency laser beam can excite the medium in the cascade way and with owing the selection rules, downward decay transitions also takes place in the cascade way.

The absorption takes place through three stimulated absorption cross-sections (σ_{ij}, $j=i+1$) in the cascade way and the downward transitions carried through stimulated emission cross-sections (σ_{ji}, $j=i+1$) and decay time of energy levels (τ_{ji}, $j=i+1$). According to Einstein stimulated excitation theory, stimulated absorption cross-section is equal to the stimulated emission cross-section i.e., $W_{ij}=W_{ji}$ [22]. Thus the rate equations have the form as

$$n_0 + n_1 + n_2 + n_3 = 1 \tag{1a}$$

$$\frac{dn_0}{dt} = -W_{01}(n_0 - n_1) + \frac{n_1}{\tau_{10}} \tag{1b}$$

$$\frac{dn_1}{dt} = W_{01}(n_0 - n_1) - W_{12}(n_1 - n_2) - \frac{n_1}{\tau_{10}} + \frac{n_2}{\tau_{21}} \tag{1c}$$

$$\frac{dn_2}{dt} = W_{12}(n_1 - n_2) - W_{23}(n_2 - n_3) - \frac{n_2}{\tau_{21}} + \frac{n_3}{\tau_{32}} \tag{1d}$$

$$\frac{dn_3}{dt} = W_{23}(n_2 - n_3) - \frac{n_3}{\tau_{32}} \tag{1e}$$

The rate Eqs. (1a)–(1e) are numerically solved and obtained the populati|1⟩on in each energy level [21]. The absorption coefficient is $\alpha_{eff}=\alpha_0(n_0\text{-}n_1)+\alpha_1(n_1\text{-}n_2)+\alpha_2(n_2\text{-}n_3)$. Here, $\alpha_0=N\sigma_{01}$, $\alpha_1=N\sigma_{12}$, and $\alpha_2=N\sigma_{23}$ are respective linear absorption coefficients from $|0\rangle$, $|1\rangle$, and $|2\rangle$ energy levels to their next higher energy levels.

The lifetime's effect on the transmittance curve is shown in Fig. 2. In four level absorption process, the transmittance get saturates

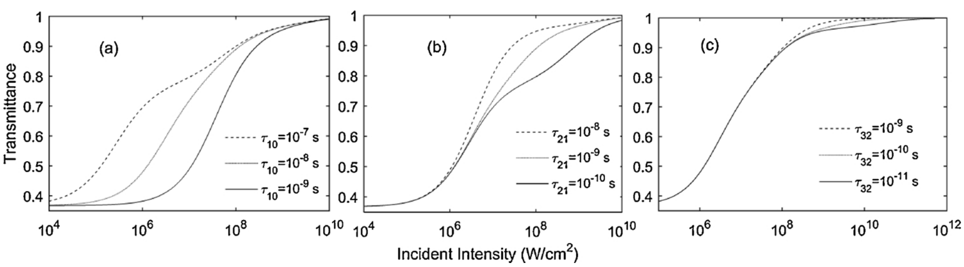

Fig. 2. Transmittance curves as a function of lifetimes: $\tau_{10} = 10^{-8}$ s (variable only in a), $\tau_{21} = 10^{-9}$ s (variable only in b), $\tau_{32} = 10^{-10}$ s (variable in only c), $\sigma_{01} = 10^{-17}$ cm^2, $\sigma_{12} = 5 \times 10^{-18}$ cm^2, and $\sigma_{23} = 10^{-18}$ cm^2.