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Comparison of rate equation models for nonlinear absorption

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ABSTRACT

Estimation of optical properties of the material depends not only on the experimental conditions, but also on the theoretical models used in the experimental data interpretation. This paper has focused on the rate Equations for two, three and four level models and solved the rate Equations analytically to interpret the effect of population dynamics in the nonlinear absorption process. In subsequent years we can see that researchers are using two rate Equation models to study the nonlinear absorption. In this article, the two models are comparatively studied under similar conditions and pictorially shown how these two models deviate from each other for same absorption cross-section and life-time parameters. The derived analytical solutions can be useful to the interested researchers in the lasers and nonlinear optics fields.

1. Introduction

Rate Eq. models are the best and the easiest way to understand the population dynamics of the energy levels of the materials [1]. Nonlinear absorption has been given a new window for nonlinear optics in the application point of view [2–7] and it is a well-established field. The nonlinear absorption properties of materials can be extracted by simple z-scan experiment [8]. In the recent years, all material scientists are using this technique for their material characterization [9–12]. Even though the technique is simple, but theoretical models used to extract the material properties are somewhat tricky. When more than two energy levels participated in the absorption process, then there is only one practical way to analyze the experimental data is by rate Eq. approach. In the process of experimental data analysis through the rate Eqs., it is essential to understand their basic criteria in the experimental data interpretation.

Rate Eqs. are using in two ways to extract the absorption cross-sections and life-times of the materials. The first one is by considering the pumping rate and absorption coefficient as a function of population difference of transition energy levels [13–20] and the second one is by considering the pumping rate and absorption coefficient as a function of the population of the ground energy level of transition [21–32]. Due to the difference in their considerations, the estimated life-times and absorption cross-sections from both the models under similar experimental conditions will be different. In this article, we have solved the rate Eqs. analytically and obtained the population in each energy level. The parameters used in the calculations: excitation wavelength (λ) is 532 nm, the thickness of the sample (L) is 0.1 mm and number density of absorption species (N) interacting with the laser beam is 10^{19} absorption species/cm^3. The number density of molecules in each state $|i\rangle$ is considered to be N_i with satisfying the condition $N = \Sigma N_i$ and the fractional number density of each state is $n_i = N_i/N$, with $\Sigma n_i = 1$. The pumping rate from $|i\rangle$ to $|j\rangle$ level is $W_{ij} = (I/h\nu)\sigma_{ij}$, where $\sigma_{ij}$is absorption cross-section from $|i\rangle$ to $|j\rangle$ level.

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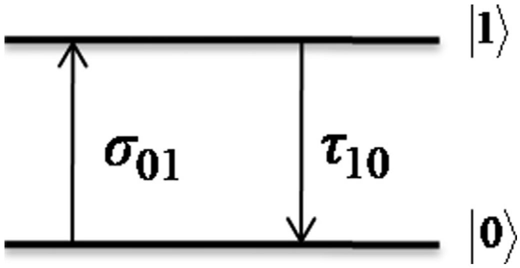

Fig. 1. Two level system.

The decay time from $|j\rangle$ to $|i\rangle$ level is τ_{ji}. Is_{ij} is saturation intensity of $|j\rangle$ state with respect to $|i\rangle$ state. The saturation of an excited state depends on the transitions between the excited state and pumping state. These transitions controlled by the excited state radiative decay life-time to ground state and absorption cross-section from ground state to excited state. In all the models, we have solved the rate Eqs. under steady state condition, where the rate of change of population with respect to time will be zero i.e., $dn_i/dt = 0$. The transmittance of medium is given by $T = \exp(-\alpha_{eff}L)$, where α_{eff} is effective absorption coefficient and depends on the number of energy levels involved in the absorption process.

2. Two level model

As shown in Fig. 1, two energy levels participate in the absorption process and the population transition between the energy levels takes place through a single absorption cross-section and fluorescence life-time.

2.1. Model 1

The rate Eqs. of population dynamics are

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

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

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

The first Eq. is the constraint and other two Eqs. give the population dynamics. Eqs. (1b) and (1c) are same and we have to acquire two variables n_0 and n_1. By solving either of Eqs. (1b) and (1c) with Eq. (1a), we can obtain the population in the ground and excited state energy levels as

$$n_0 = \frac{1 + \left({}^{I_{S01}}\!/_{I}\right)}{2 + \left({}^{I_{S01}}\!/_{I}\right)} \tag{2a}$$

$$n_1 = \frac{1}{2 + \left({}^{I_{S01}}\!/_{I}\right)} \tag{2b}$$

The absorption coefficient is given by

$$\therefore \alpha_{eff} = N\sigma_{01}(n_0 - n_1) = \alpha_0 \frac{\left({}^{I_{S01}}\!/_{I}\right)}{2 + \left({}^{I_{S01}}\!/_{I}\right)} \tag{3}$$

$I_{S01} = h\nu/(\sigma_{01}\tau_{10})$ is saturation intensity of excited state $|1\rangle$ with respect to ground state $|0\rangle$ and the linear absorption coefficient $\alpha_0 = N\sigma_{01}$.

2.2. Model 2

The rate Eqs. of population dynamics are

$$n_0 + n_1 = 1 \tag{4a}$$

$$-W_{01}n_0 + \frac{n_1}{\tau_{10}} = \frac{dn_0}{dt} \tag{4b}$$

$$W_{01}n_0 - \frac{n_1}{\tau_{10}} = \frac{dn_1}{dt} \tag{4c}$$