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Wavelength dependent resonant nonlinearities in a standard saturable absorber IR26 on picosecond time scale

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ABSTRACT

Nonlinear optical properties of a standard dye IR26 have been studied by using the Z-scan technique to decipher the difference in the mechanism of nonlinear absorption on picosecond time scale at two wavelengths i.e. at 1064 nm and 532 nm. A prominent contribution of nonlinear absorption is observed in the Z-scan profiles at 1064 nm. The dye exhibits the mechanism of self-defocusing at 1064 nm in contrast to that of self-focusing at 532 nm. While the two photon absorption has been found to be the dominant mechanism of reverse saturable absorption at 1064 nm, the mechanism of excited state absorption is operating at 532 nm. Additionally, the optical phase conjugate geometry of degenerate four wave mixing (DFWM) technique has been used to measure the third order nonlinear susceptibility values at 532 nm to compare with those obtained from the Z-scan profiles.

1. Introduction

Heptamethine cyanine dyes are an important class of chromophores with absorption and fluorescence spectra in visible and near infrared regions [1]. Due to their high relative photostability and high molar extinction coefficient, these have applications in various fields, such as modelocking in lasers [2], fluorescent labeling for proteins [3], fluorescent tags in DNA sequencing [4], immunoassay [5], flow cytometry [6], and in optical recording medium [7]. Several IR dyes were investigated more than two decades back [8]. A series of distinguished multifunctional chromophores, cyanine dyes and their J and H-aggregates [9] have recently evoked interest in the areas of optoelectronics and nonlinear optics. Even though one of the standard heptamethine cyanine dye IR26 has been used for passive modelocking in commercial lasers [10], its nonlinear optical properties have not been studied so far.

Z-scan technique [11] is used to measure the nonlinear refractive (NLR) and nonlinear absorptive (NLA) parameters of a material in the time scales of fs [12] to ms [13]. This single beam measurement technique not only gives the sign but also the magnitude of the nonlinearity. The technique is based on the principle of spatial beam distortion of the Gaussian beam at the exit surface of the sample. Due to presence of the space dependent refractive index changes, the self-focusing and self-defocusing effects can be observed. On the other hand, refractive index changes due to the absorptive nonlinearity give rise to the phenomena of saturation absorption (SA) and reverse saturable absorption (RSA).

An economical alternate for the semiconductor saturable absorber mirror technology [14] can be dye-embedded solid-state saturable absorber devices in the IR region. In this direction we have found a new class of highly efficient Weinreb amide based dyes [15]. To extend these studies in IR region, we have studies here a standard dye IR 26 as, to the best of our knowledge, its nonlinear optical parameters are not reported till date. It is found that the dye has negative NLR and a high value of negative nonlinear absorptive index at 1064 nm. In contrast, it shows positive NLR and positive nonlinear absorptive index at 532 nm. While the SA is the main mechanism operating at 1064 nm, the Z-scan profiles are dominated with the RSA at 532 nm. The irradiance dependence of the Z-scan profiles at the two different wavelengths are also given here. The optical phase conjugation (OPC) has been used to measure the third order nonlinear susceptibility ($\chi^{(3)}$) values at 532 nm to compare those obtained from the Z-scan profiles.

2. Experimental

The dye IR26 (Lambdachrome) and the solvent 1,2-dichloroethane (Aldrich) were used as received. The absorption spectrum (Fig. 1) was recorded using a UV–visible spectrometer (JASCO, V-570) in quartz cell of 1 mm thickness. The experimental arrangements for the Z-scan and OPC techniques are given in Fig. 2. The fundamental (1064 nm) and the second harmonic (532 nm) wavelengths of a picosecond Nd: YAG laser (Continuum Model YG601, 40 ps, 10 Hz) were used for the Z-scan experiments. The transmitted energy as a function of the sample position at the far field was measured with the help of the

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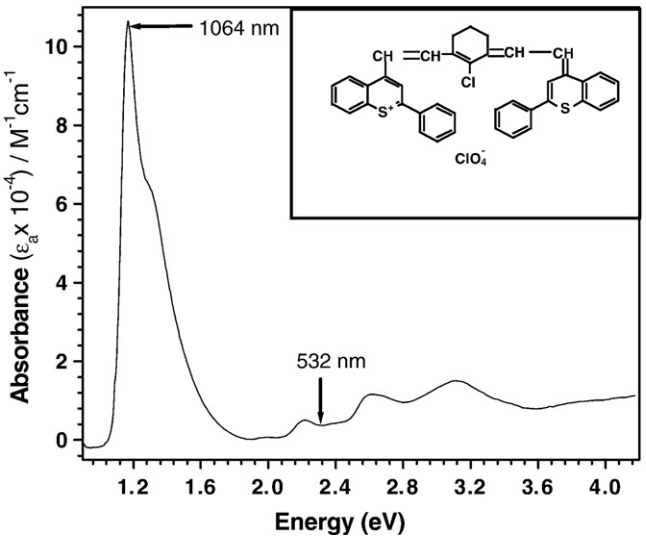

Fig. 1. The absorption spectrum of the dye IR26 in 1, 2, dichloroethane. The arrows indicating the pump wavelengths. The inset shows the molecular structure of the dye.

photodiode (Becker and Hickle, PDI-400) after focusing with a double convex lens of focal length of 50 mm. A circular aperture of 1 mm diameter is used before the detector at the far field for the closed aperture (CA) Z-scan experiments. A large diameter double convex lens of focal length 200 mm replaced the circular aperture for the open aperture (OA) Z-scan. The beam was nearly Gaussian and we have considered the 86% beam criterion [16]. The radius of the beam waist (ω_o) was 24 μm with the corresponding Rayleigh range (z_R) of 3.5 mm. The sample thickness (1 mm) was less than the Rayleigh range to fulfill the thin sample approximation condition. In case of OPC, the incident beam was split into three beams (two pumps and one probe beam). Two strong-counter propagating pump beams create the grating, while the weaker probe is used for detecting the phase conjugation (PC) signal. The angular separation between the forward pump and the probe is about 8° at the sample. The phase conjugate beam that retraces the path of the probe was separated by placing a beam splitter. The signal detected by the photodiode was averaged over hundred shots using a 100 MHz digital oscilloscope (Tektronics TDS220).

3. Theory

A general analysis of a saturable absorber is given by Hercher [17] using the three-level model. Under the steady state approximation, where the population of energy levels remains constant for the duration of the pulse, a general expression for the absorption coefficient in terms of saturation intensity (I_S) is given by

$$\alpha = \frac{\alpha_o}{1 + I/I_S}$$

Where $I_S = (h\nu/\sigma_o\tau)$ is the saturation intensity, $h\nu$ is the photon energy, and τ is the lifetime of the excited state. I_S is the intensity at which the absorption coefficient drops to the half of its linear value. In RSA the transmittance decreases on increasing the input intensity due to excited state absorption (ESA) or two photon absorption (TPA). In saturable absorption (SA) however, the transmission increases at higher incident intensities, indicating the further bleaching of the ground state due to ESA.

To discuss the various parameters that influence the RSA and the SA a general model of three-level energy diagram [18–20] is used (Fig. 3). Here S_n (n = 1,2,...) are singlet excited states while S_o is the singlet ground state. The molecules get to the excited state from the ground state by absorbing the incident laser photons. These excited molecules relax to the S_1 state in ps time scale by nonradiative decay. The excited molecules may also make transitions to the triplet state (not shown here) with the mechanism of intersystem crossing within the time scales of ns. The molecules in S_1 can be further excited to the higher energy state S_2. This process is known as the excited state

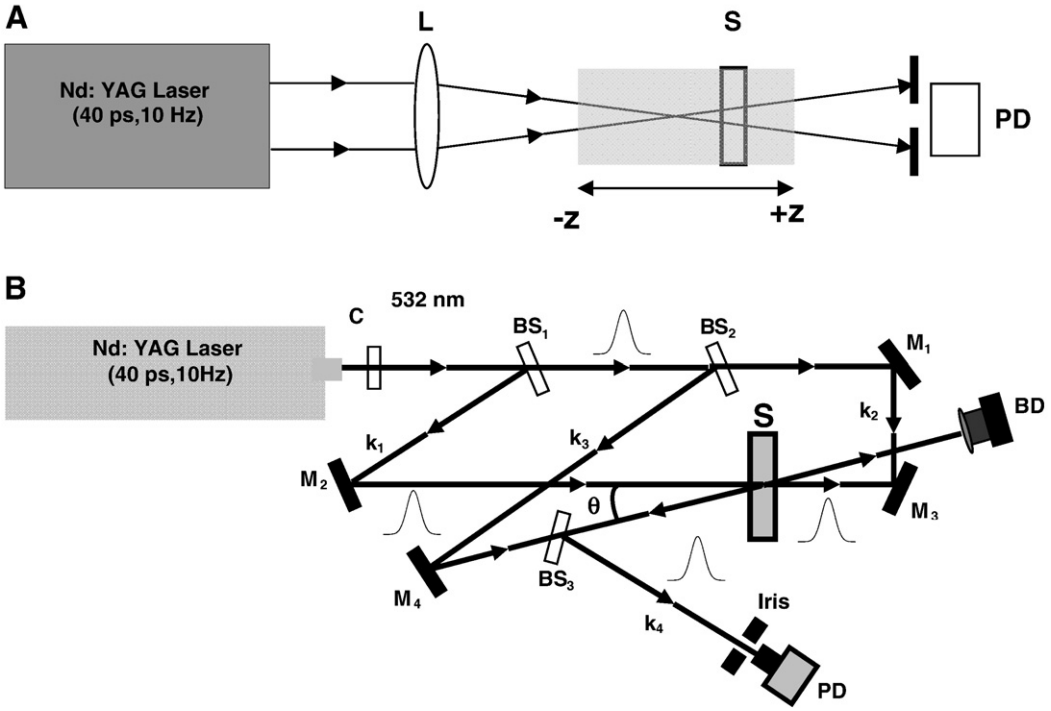

Fig. 2. The experimental setup (A) for Z-scan, L—lens, S—sample, PD—photodiode and (B) for optical phase conjugation. BS1, BS2, BS3 are beam splitters and M1, M2, M3, and M4 are the mirrors.