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Z-scan determination of the third-order optical nonlinearity of a triphenylmethane dye using 633 nm He–Ne laser

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Abstract

The third-order nonlinear optical response of a triphenylmethane dye (Acid blue 7) was studied using the Z-scan technique with a continuous-wave He–Ne laser radiation at 633 nm. The magnitude and sign of the third-order nonlinear refractive index n_2 of aqueous solution of Acid blue 7 dye were determined; the negative sign indicates a self-defocusing optical nonlinearity in the sample studied. The negative nonlinear refractive index n_2 and nonlinear absorption coefficient β were estimated to be -1.88×10^{-7} cm^2/W and -3.08×10^{-3} cm/W, respectively, corresponding to $\mathrm{Re}(\chi^{(3)}) = -8.35 \times 10^{-6}$ esu, and $\mathrm{Im}(\chi^{(3)}) = -6.88 \times 10^{-7}$ esu. The experimental results show that Acid blue 7 dye have potential applications in nonlinear optics.

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Keywords: Nonlinear refractive index; Nonlinear absorption; Triphenylmethane dyes; Z-scan technique

1. Introduction

The nonlinear properties of organic dye molecules are currently being explored with great interest [1–3]. The nonlinear refractive index is a very important parameter in designing optical devices. Previous measurements of nonlinear refraction have used a variety of techniques including nonlinear interferometry [4,5], degenerate four-wave mixing [6], nearly degenerate three-wave mixing [7], ellipse rotation [8] and beam distortion measurements [9]. The first three methods, namely nonlinear interferometry and wave mixing are potentially sensitive techniques, but all require relatively complex experimental apparatus. Beam distortion measurements, on the other hand, are relatively insensitive and require detailed wave propagation analysis. The Z-scan technique is based on the principles of spatial beam distortion and offers simplicity as well as very high sensitivity for measuring both the nonlinear refractive index and nonlinear absorption coefficient. Z-scan technique, originally proposed by Sheik-Bahae et al. [10,11] has been since then implemented and applied to the study of several nonlinear composite glasses and semiconductor materials [12,13]. Nonlinear optical properties of organic dye molecules are important from the point of view of understanding their photo-physics and for realizing the potential for using these molecules in applications like optical processing, computing [14] and optical limiting [15].

In this work, we report the experimental measurements of the third-order nonlinear refractive index n_2, absorption coefficient β and the nonlinear susceptibility ($\chi^{(3)}$) of an organic dye (Acid blue 7) using Z-scan technique with a continuous-wave He–Ne laser radiation at 633 nm. We carried out both closed and open aperture Z-scan measurements in aqueous solution of Acid blue 7 dye at an input intensity of 2.39 kW/cm^2. The experiments were repeated for different dye concentrations and the third-order nonlinear refractive index is found to be linearly dependent on the dye concentration with in the range studied.

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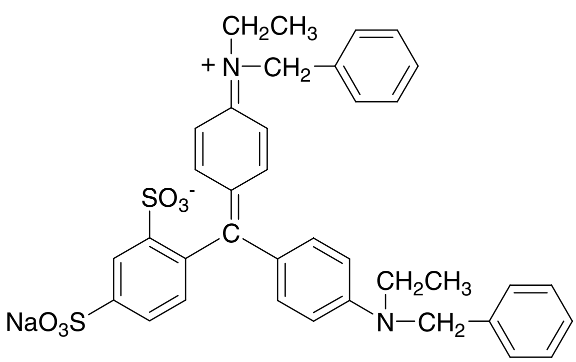
2. Materials

The general structure and formula of the triphenylmethane dye Acid blue 7 (Alphazurine A) are illustrated in Fig. 1. The chromophore of this dye is the quinonoid group, which appears as $C = ArNR_1R_2$ and it is a diamino derivative of the triphenylmethane group. Acid blue 7 is in the form of a dark bluish powder and is highly soluble in water giving a bluish-green solution. The UV–visible absorption spectra of Acid blue 7 dye were studied using UV-2401 PC Spectrophotometer. The optical absorption of the Acid blue 7 dye in water with 0.01 mM concentration shows an absorption peak (λ_{max}) at 637 nm as shown in the Fig. 2.

3. Experimental studies

The Z-scan technique is a simple and effective tool for determining the nonlinear properties of various kinds of materials because it provides not only the magnitude of the real and imaginary parts of the third-order nonlinear susceptibility $\chi^{(3)}$, but also the sign of the real part. Nonlinear index of refraction n_2 is proportional to the real part of the third-order susceptibility ($\mathrm{Re}\{\chi^{(3)}\}$) and the nonlinear absorption coefficient β is proportional to the imaginary part of the third-order susceptibility ($\mathrm{Im}\{\chi^{(3)}\}$).



Molecular formula: $C_{37}H_{35}NaN_2O_6S_2$

Fig. 1. Chemical structure and formula of Acid blue 7 dye.

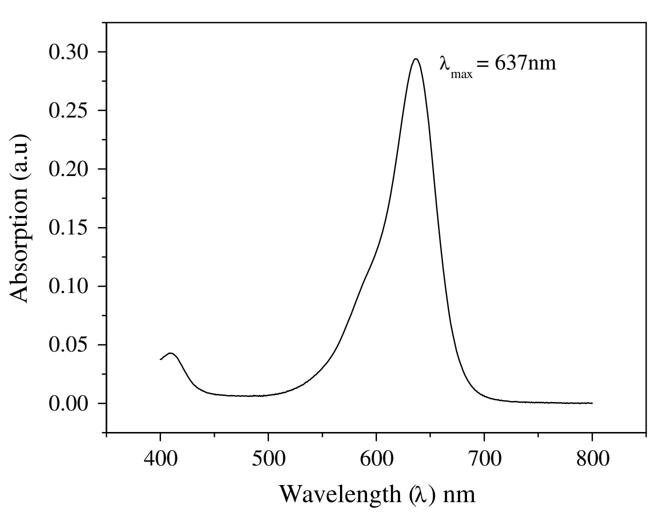


Fig. 2. UV–Vis absorption spectra of Acid blue 7 dye in water.

In our experiment, a plane polarized Gaussian laser beam, propagating in the z-direction, is focused to a narrow waist. The sample is translated along the z-direction and the transmitted intensity is measured through a finite aperture in the far field as a function of the sample position z, measured with respect to the focal plane. As the sample moves through the beam focus (at $z = 0$), self-focusing or defocusing modifies the wave front phase, thereby modifying the detected beam intensity. The schematic set-up of the Z-scan technique is shown in Fig. 3.

To figure out how the Z-scan transmittance as a function of z is related to the nonlinear refraction of the sample, let us assume a medium with negative nonlinear refraction index and thickness smaller than the diffraction length of the focused beam. This can be considered as a thin lens of variable focal length. Beginning far from the focus ($z < 0$), the beam irradiance is low and nonlinear refraction is negligible. In this condition, the measured transmittance remains constant (i.e., z-independent). As the sample approaches the beam focus, irradiance increases, leading to self-lensing in the sample. A negative self-lens before the focal plane will tend to collimate the beam on the aperture in the far field, increasing the transmittance measured at the iris position. After the focal plane, the same self-defocusing increases the beam divergence, leading to a widening of the beam at the iris and thus reducing the measured transmittance. Far from focus ($z > 0$), again the nonlinear refraction is low resulting in a transmittance z-independent. A pre-focal transmittance maximum (peak), followed by a post-focal transmittance minimum (valley) is a Z-scan signature of a negative nonlinearity. An inverse Z-scan curve (i.e., a valley followed by a peak) characterizes a positive nonlinearity.

The Z-scan experiments were performed using a 633 nm, He–Ne laser beam (35 mW, Coherent, 31-2140-000), which was focused by 3.5 cm focal length lens. The laser beam waist ω_0 at the focus was measured to be 21.28 μm and the Rayleigh length $z_R = 2.24$ mm. A 1 mm thick cuvette (cell) containing the aqueous solution of Acid blue 7 dye was translated along the axial direction that is the direction of the propagation of the laser beam. The intensity of the

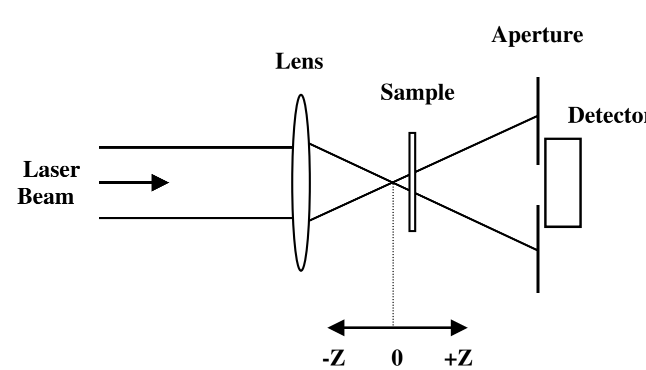


Fig. 3. Schematic diagram of experimental arrangement for the Z-scan measurement.