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χ(3) – based properties of 3,4-dimethoxydibenzylideneacetone by the single beam Z-scan method

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ABSTRACT
Because of its many uses in photonics and optoelectronics, nonlinear optics is gaining the interest of researchers all over the world. It is required to look into the big nonlinear optical properties of new or current materials in order to construct devices for such applications. Due to their propensity to crystallise in noncentrosymmetric structures and their ability to transmit blue light, chalcones have attracted a great deal of interest as materials for second-order nonlinear optical applications. Chalcones can, however, have significant third-order nonlinearities because of their -conjugated structure and their nature as charge transfer chemicals. In this article, we present the results of a single beam Z-scan examination into the third-order nonlinear optical (TNLO) characteristics of a chalcone derivative, specifically 3,4-dimethoxydibenzylideneacetone. We discovered that adding electron-donating groups to the fundamental molecule dibenzylideneacetone can improve the characteristics of TNLOs. It is discovered that the nonlinear refractive index is negative and has a magnitude of 10-10 esu. At 532 nm, 3,4-dimethoxydibenzylideneacetone shows significant two-photon absorption. With this compound, I show effective optical limiting of nanosecond laser pulses. The research demonstrates the appeal of 3,4-dimethoxydibenzylideneacetone for optical limiting applications.

1. Introduction

Owing to the important usage in all optical switching systems, including optical modulations and the optical limiting of lasers, nonlinear media exhibiting significant cubical nonlinear optical susceptibility ($\chi^{(3)}$) are recently of significant interest [1-3]. Particularly in optical limiting applications utilising nanosecond, materials with significant two-photon absorption coefficients are becoming increasingly important. Large two-photon absorption cross-section organic conjugated media having donor-acceptor-donor formation are synthesized [4]. Novel semiconductors and organic materials have received a lot of attention recently since nonlinear optical materials have developed significantly. Since their molecular nature and the adaptability of synthetic chemistry can be used to change and optimise molecular structure to maximise third order nonlinear responses and alter properties [4, 5], organic molecular media are progressively more being recognised as the future media among these materials. Organic molecules display the biggest non-resonant (non-absorptive) optical nonlinearities due to their distinctive chemical structure (bonding). In these materials, the significant electron delocalization leads to extremely high molecule polarizability and impressive third order nonlinearities [6]. Optical limiting, or the rise in optical absorption with light intensity, is the most potential use of nonlinear optical activity. For the purpose of optically restricting nanosecond and picosecond laser pulses, materials with substantial two-photon absorptivities are being researched [7, 8]. In the current work, we provide the results of an experimental study employing nanosecond laser pulses at 532 nm wavelength to examine third order optical characteristics of 3,4-dimethoxydibenzylideneacetone. We discuss the impact of methoxy group substitution on the dibenzyledeneacetone's nonlinear response. We also discuss the compound's optical limiting (OL) and two-photon absorption (TPA) characteristics.

2. Objectives

In order to determine the potential for OL applications, the goal of present research is to investigate the third-order optical susceptibility ($\chi^{(3)}$) of a chalcone derivative, specifically 3,4-dimethoxydibenzylideneacetone.

3. Experiment

The sample's nonlinear susceptibility was measured via single beam Z-scan method [9]. The Z-scan experiment made use of a Q-switched Nd:YAG laser emitting optical pulses of width 7 nano-second operating at 532 nano-meter. Gaussian beam was utilised in the Z-scan technique. The laser pulse was focused onto the 1 milli-metre quartz cavetti, which held the solution of the sample, using a convex-lens having the 26 centi-meter focal length. At focused spot, the laser beam's waist radius was measured to be 19.6 μm. It is equivalent to 2.274 milli-meter Rayleigh's length. As a result, since the width of 1 milli-meter of the sample was smaller in comparison to Rayleigh's length, it was considered a thin film. The scan's 50% (S = 0.5) aperture and pulse energy of

0.35 milli-joule resulted in the peak irradiance of 7.3 GW-cm⁻², respectively. The focus was 30 centi-meter away from the limiting aperture. In order to prevent the accumulating thermal effects, value was gathered using single shot approach [10]. In the OL experiment, a 26 centi-meter convex lens focused the coherent radiation to the centre of the quartz cell with a 1 milli-meter path length, resulting in a focal point with a $1/e^2$ radius of around 19.6 meter. During the experiment, the medium was positioned at the focus position. By changing the input pulse power by rotating a polarizer and measuring output as well as input power/energy by making the use of two 13-PEM-001 series Melles Griot power/energy meters, optical limiting was achieved (detectors).

The Ocean Optics SD 2000 type UV-VIS fibre optic spectrometer was used to record the (linear) absorbing spectrum of DBA mixed in dimethylformamide (DMF) in a 1-centi-meter size quartz cell. The Abbe refractometer was inserted to calculate compound's (linear) index of refraction.

4. Results and discussion

According to the accepted method described in the literature [11], p-methoxydibenzylideneacetone was produced in the materials science lab of DCRUS&T, Murthal, Haryana.
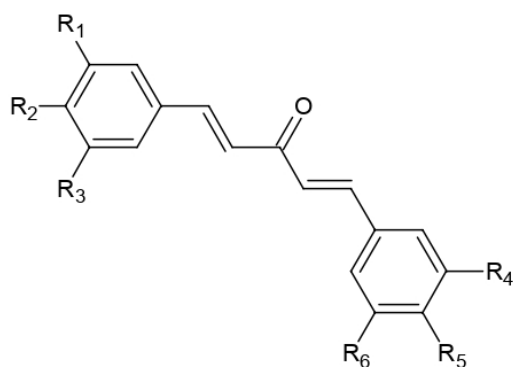

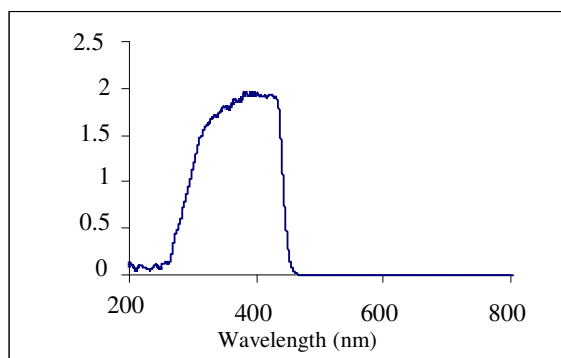**Figure 1.** Structure of 3,4-dimethoxydibenzylideneacetone. R_2, R_3, R_5, R_6 = OCH_3; R_1 and R_4 = H.

Figure 2. Linear Absorption Spectrum of sample dissolved in DMF (1×10^{-2} mol/L).

Figure 1 depicts the p-methoxydibenzyledeneacetone crystal structure. Figure 2 depicts the compound's linear absorption spectrum. The sample's UV-VIS absorption spectrum was captured in diluted solutions (1×10^{-2} mol/L) at room temperature. Since the material is transparent at 532 nm according to the spectra, we may quantify non-resonant optical nonlinearity.

As the sample passed through the focus point, the nonlinear transmission of compounds with and without an aperture was evaluated in the far field. As a result, we can distinguish between nonlinear refraction and nonlinear absorption. Figure 3 (a) and (b), respectively, depict the samples' pure nonlinear refraction and open aperture refraction curves. The normalised transmission at 532 nm without an aperture is depicted in Figure 3(a). The transmission in this case exhibits an intensity-dependent absorption effect and is symmetric in connection with the focus (z = 0), it possess a negligible transmittance. Experimental data are perfectly matched by the equation provided in Ref. [9]. TPA characteristics of the compound have been indicated in form of the open aperture curve [12]. To calculate samples' nonlinear absorption coefficient (β), the model mentioned in Ref. [9] was utilised.
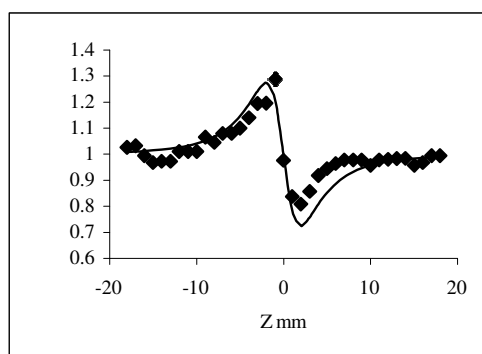

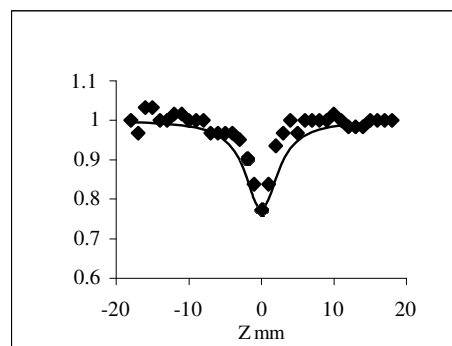**Figure 3.** (a) Normalized open aperture curve for 3,4-dimethoxydibenzylideneacetone.

Figure 3. (b) Normalized closed aperture curve. Solid lines are fits of data to equations given in Ref. [9].

The normalized transmittance of chemicals via closed aperture has been depicted in Figure 3(b). The division method was applied to generate this pure nonlinear refraction curve. Strong refractive nonlinearity can be seen as a pronounced peak in the Z-scan curve. Additionally, the closed aperture curve's peak-valley structure amply demonstrates a negative refractive nonlinearity. Repeating the Z-scan on each sample yielded the magnitudes of n_2, Im $\chi^{(3)}$ and Re $\chi^{(3)}$ reported in Table 1, and the values were constant across all trials.

The pure nonlinear refraction curve's peak-to-valley shape in Figure 3(c) makes it abundantly evident that the response is electronic in origin, and change in sign of refractive index is negative, demonstrating that the heating effect is not the solution's primary source of third-order nonlinear response. Additionally, the author performed a z-scan technique on pure

DMF to investigate the solvent's contributions to n_2, and the author discovered that the nonlinear absorption/refraction were not seen at an input energy employed. As a result, the solvent barely contributes to the sample's nonlinearity.

Table 1. The calculated values of n_0, n_2, Re $\chi^{(3)}$ and Im $\chi^{(3)}$ for 3,4-dimethoxydibenzylideneacetone

n_0	n_2 (e.s.u.)	β (cm/GW)	Re $\chi^{(3)}$ (e.s.u.)	Im $\chi^{(3)}$ (e.s.u.)
1.424	-2.206×10^{-10}	3.90	-2.376×10^{-13}	0.604×10^{-13}

The magnitude of $\chi^{(3)}$ may be increased by constructing a donor-acceptor-donor structure to enhance the extent of transfer of charge from the ends of molecules to the centre [4]. To boost the response, we therefore substituted methoxy group, an electron donor, in the 3 and 4 positions of dibenzylideneacetone. Comparing the response to dibenzylideneacetone, which has a $\chi^{(3)}$ of -0.38×10^{-13} esu, it was discovered that the response substantially doubled [2]. The enhanced delocalized electron density in the molecule is what causes this improvement in responsiveness.

At 532 nano-meter, the molecule exhibits extremely potent TPA. For nano-second pulsed lasers, a strong optical limitation based on TPA was seen. Figure 4 depicts the optical limiting of nanosecond pulses in 3,4-dimethoxydibenzylideneacetone.
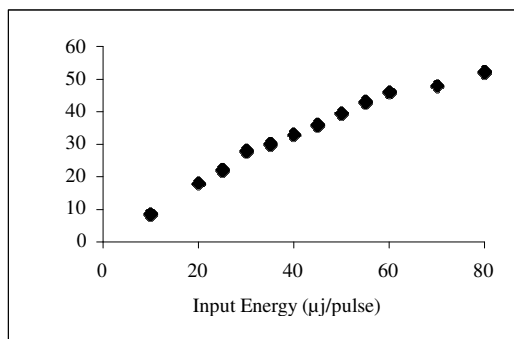

Figure 4. Optical limiting of nano-second pulses in 3,4-dimethoxydibenzylideneacetone

For an input energy less than 55 µJ/pulse, the output energy depicts a linear increment with incident energy. However, the optical limitation was severed over the 55 µJ/pulse. The author could not detect the considerable nonlinear scattering in the solution during the Z-scan experiment, despite the fact that nonlinear absorption and nonlinear scattering can both contribute to the optical limitation. This shows that the effective optical limiting performance is mostly caused by the substantial two-photon absorption. This demonstrates that the substance is a suitable substance for optically restricting nanosecond laser pulses.

5. Conclusions

Using 7 nano-second laser pulses at 532 nano-meter, the Z-scan and nonlinear transmission studies were carried out on 3,4-dimethoxydibenzylideneacetone. The author calculated the compound's n_2, Re $\chi^{(3)}$ and Im $\chi^{(3)}$ values. The nonlinearity of dibenzylideneacetone increased when a methoxy group was substituted, according to Z-scan data, and this indicates that the

response is electronic in nature. The substance has a weak two-photon absorption at the applied wavelength and a negative nonlinear refractive index. Studies on the compound's nonlinear transmission show that TPA helps to cause the OL behaviour. According to the investigations, the substance has a good chance of being used in photonic and optoelectronic applications.

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