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Third-Order Nonlinear Optical Properties of 1,3-Bis(3,4-Dimethoxyphenyl) prop-2-en-1-one Under Femtosecond Laser Pulses

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Abstract. In this paper, we present the third-order nonlinear optical (NLO) studies of1,3-bis(3,4-dimethoxyphenyl)prop-2-en-1-one (abbreviated as VDMC). The chalcone was synthesized by Claisen-Schmidt condensation method. The third-order nonlinear optical properties were evaluated using standard, well-known Z-scan technique under femtosecond laser regime (150 fs, 900 nm) with two different laser repetition rates 500 Hz and 80 MHz. Open aperture studies showed that the molecule possess two photon absorption with the coefficients in the order 10^{-9} cmW^{-1}. The closed aperture studies have resulted the negative nonlinear refraction with the coefficients in the order 10^{-14} cm^2W^{-1}. The two-photon absorption cross sections were estimated. Optical limiting properties have been studied and the limiting threshold values were found to be in the range 0.86-2.3 mJ/cm^2, which suggests that VDMC has better applications in the field of nonlinear optics.

1. Introduction

Recently, nonlinear optical (NLO) and optoelectronic materials have got the researchers attention due to their advanced applications in display technologies, laser industry, and telecommunications[1]. As an interesting type of nonlinear optical materials, the chalcones (1, 3-diaryl-2-propen-1-ones) have been received much attention due to their notable second and third-order non-linear optical responses[2, 3]. Interestingly, these types of compounds exhibit the enhancement of optical nonlinearities attributed to the introduction of electron donor and electron acceptor on either side of aromatic rings to the large π-conjugated system, strong intermolecular interaction, as well as 2PA resonance[4, 5]. In the present study, we synthesized a chalcone 1,3-bis(3,4-dimethoxyphenyl)prop-2-en-1-one (VDMC) and crystals were grown under slow evaporation solution growth technique. The third-order nonlinear optical properties were investigated by using Z-scan technique under femtosecond (fs) laser regime at 900 nm wavelength and 500 Hz and 80 MHz chopper frequencies. The two-photon absorptions, negative nonlinear refractions and the optical limiting studies have been highlighted in the current work for the VDMC title molecule.

2. Experimental

VDMC compound was synthesized by using the Claisen-Schmidt condensation reaction method[6]. Here the 3,4 dimethoxyacetophenone was treated with 3,4-dimethoxybenzaldehyde in ethanol in the presence of a catalytic amount of NaOH solution. After stirring for 5h, the contents of the flask were poured into ice-cold water and left to stand for 1h. The resulting crude solid was filtered, washed successively with dilute HCl solution and distilled water, and finally recrystallized repeatedly from acetone to give the pure VDMC. The chemical structure of the compound is shown in **Fig 1.** The UV-Vis absorption studies suggests that the VDMC molecule has optical transparency in the region 450-1000 nm.

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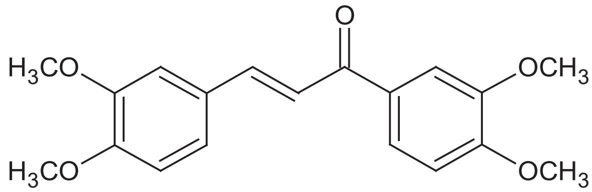


Fig 1. Molecular structure of VDMC.

The nonlinear optical (NLO) properties of VDMC were measured by using standard Z-Scan technique[7] under femtosecond laser regime (900 nm, 150 fs) at two different laser repetition rates (500 Hz, 80 MHz). Z-scan technique has great advantages due to its simplicity and high sensitivity; it can simultaneously measure both sign and magnitude of nonlinear refraction and nonlinear absorption. In this technique, the sample was scanned through the focal plane of a tightly focused Gaussian beam. Meanwhile, the changes in the far-field intensity pattern with and without aperture are monitored. Experiments were performed using fs pulses from a Ti: sapphire oscillator (maximum average power of ~4 W). The Z- scan experiments are done with typically 20-30 mW input power with corresponding pulse energy being ~50 nJ. The pulses were tunable in the wavelength region of 680-1060 nm. The sample was scanned along the Z – direction through the focus of the beam passed through a 100 mm focal length lens. The input beam was spatially filtered to attain a pure Gaussian profile in the far field. The sample was placed on a 10 µm resolution translation stage and the data was collected manually using power meter detector (Field-Max, Coherent). The transmitted intensity was recorded as a function of the sample position. The beam waist (ω_0) estimated at the focus was ~25 µm with a corresponding Rayleigh range of ~2.26 mm. The closed aperture scan was performed at intensity where the contribution from the higher order nonlinear effects is negligible. The experiment was performed for two different chopper frequencies 500 Hz and 80 MHz by placing an optical chopper in the experimental setup at 900 nm wavelength. Solution of VDMC was prepared using DMF solvent with 1 mM concentration. It has been found that the results obtained from the solutions are purely due to VDMC and there is no solvent effect for NLO properties.

3. Results and discussion

3.1. Nonlinear absorption and refraction

In the Z-scan experiment, the nonlinear transmission of compounds without aperture (open aperture) measured in the far field as the sample was moved through the focal point. This allows us to determine the nonlinear absorption coefficient β. The open aperture curve of VDMC shown in **Fig 2**. Here, the transmission is symmetric with respect to focus (z = 0), where it reaches a minimum value, showing an intensity dependent absorption effect. The presence of valley in open aperture scans indicates strong reverse saturation absorption (RSA) at peak intensities. The corresponding normalized transmission as a function of sample position in open aperture condition given by[8],

$$T(Z) = 1 - \frac{\beta I_0 \, L_{eff}}{2\sqrt{2}\,(1+Z^2/Z_0^2)} \qquad (1)$$

Where, $L_{eff} = (1 - exp^{-\alpha L})/\alpha$ is the effective length of the sample, α is linear absorption coefficient and L is thickness of the sample. The two-photon absorption coefficient β is found to be 1.5×10^{-9} cmW^{-1} and 15.5×10^{-9} cmW^{-1} at 500 Hz and 80 MHz chopper frequencies, respectively.

To obtain the nonlinear refraction coefficient n_2, we fit the transmission curve by the well-established formula,

$$T(Z) = 1 - \frac{(4X\Delta\phi_0)}{[(X^2+1)(X^2+9)]} \qquad (2)$$

Where, $X=Z/Z_0$ and $\Delta\phi_0$ is the on-axis nonlinear phase shift and I_0 is the peak intensity at the focus, $k = 2\pi/\lambda$ is the wave vector. Then the third-order nonlinear refraction index n_2 can be calculated by knowing $\Delta\phi_0$ from the closed aperture Z-scan by using the relation $\Delta\Phi_0 = kn_2I_0L_{eff}$. The values of n_2 for VDMC at 500 Hz and 80 MHz chopper frequencies are found to be $-5.37\text{x}10^{-14}$ cm^2W^{-1} and $-20.18\text{x}10^{-14}$ cm^2W^{-1}, respectively.

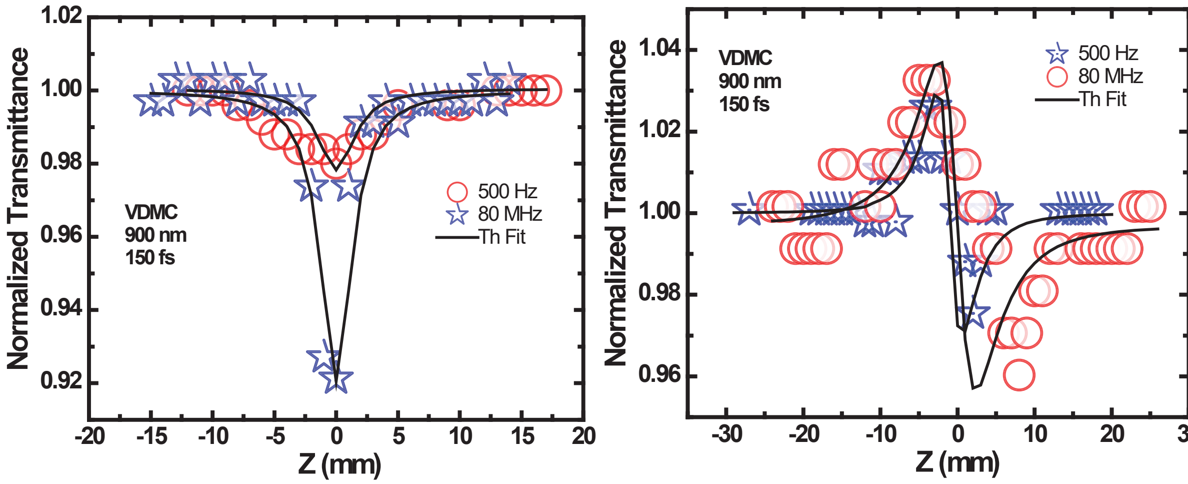
The nonlinear refractive index n_2 and nonlinear absorption coefficient β are related to the real and imaginary part of third-order nonlinear optical susceptibility $\chi^{(3)}$ through the following relations, respectively[9].

$$\chi^{(3)} = \chi_R^{(3)} + i\chi_I^{(3)} \qquad (3)$$

$$\chi_R^{(3)}(esu) = \frac{cn_0^2}{120\pi^2} n_2 \quad (m^2/W) \qquad (4)$$

$$\chi_I^{(3)}(esu) = \frac{c^2n_0^2}{240\pi^2\omega} \beta \quad (m/W) \qquad (5)$$

Where, n_0 is the linear refraction index and c is the velocity of light in vacuum, β and n_2 are the nonlinear absorption and refraction coefficients and ω is the angular frequency of the light field. The calculated values of $\chi_R^{(3)}$(esu),$\chi_I^{(3)}$(esu), total $\chi^{(3)}$(esu), molecular hyperpolarizability (γ_h) and two photon absorption cross sections (σ_{2PA}) are tabulated in the **Table 1.**



(a) (b)

Fig 2. (a) Nonlinear absorption and **(b)** refraction Z-scan curves of VDMC at two different chopper frequencies.

Table 1. Third-order nonlinear optical parameters of VDMC obtained at 1 mM solution.

Laser Rep rate	n_2 (cm^2W^{-1}) x 10^{-14}	β (cmW^{-1}) x 10^{-9}	Re χ(3) (e.s.u) x 10^{-12}	Im χ(3) (e.s.u) x 10^{-12}	χ(3) (e.s.u) 10^{-12}	γ_h (e.s.u) x 10^{-31}	σ_{2PA} (GM) x 10^4
500 Hz	-5.37	1.5	-2.75	0.61	2.81	14.54	5.5
80 MHz	-20.18	15.5	-10.32	6.33	12.1	62.6	56.8

3.2. Optical limiting

Optical limiting (OL) is a phenomena observed when the transmission of a medium decreases with increasing input laser intensity or fluence[10]. OL can be achieved through various nonlinear optical mechanisms such as multi-photon absorption (MPA), excited state absorption (ESA), free carrier absorption (FCA), self-focusing, self-defocusing, nonlinear scattering, photo-refraction etc. Coupling two or more of these mechanisms has also causes OL like self-defocusing along with MPA. Optical limiting threshold is the key feature to know whether the material is good optical limiter. It is the minimum input energy or fluence required for the laser beam to deviate the linearity of the optical limiter, there after output becomes constant (i.e. transmittance decreases with the intensity). Lower the threshold value better will be the optical limiter performance. In case of VDMC, limiting threshold was observed at 0.86 mJ/cm^2 and 2.3 mJ/cm^2 for 500 Hz and 80 MHz chopper frequencies, respectively. Beyond this value, transmission decreases dramatically with the increase in fluence as shown in Fig 3. Thus, the observed nonlinear absorption, which originates from the two-photon absorption (2PA), is responsible for the optical limiting behavior.

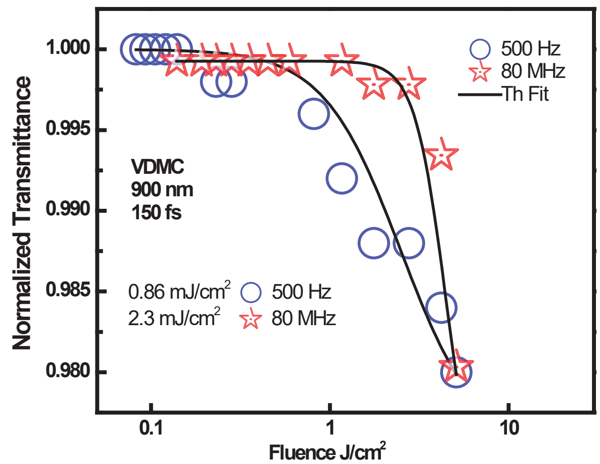


Fig 3. Optical limiting curves of VDMC.

4. Conclusions

The VDMC chalcone derivative was synthesized using Claisen-Schmidt condensation method. The third-order NLO properties were successfully studied by using Z-scan technique under femtosecond laser regime at 900 nm wavelength and 500 Hz & 80 MHz chopper frequencies. The two-photon absorptions and negative nonlinear refractions have been observed in both the laser frequencies. Further, we found good optical limiting behavior with the threshold values in the range 0.86-2.3 mJ/cm^2, which suggests that VDMC is promising candidate for possible applications in nonlinear optical devices.

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