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Study on third-order nonlinear optical properties of 4-methylsulfanyl chalcone derivatives using picosecond pulses

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ABSTRACT

In this paper we present results from the experimental study of third-order nonlinear optical (NLO) properties of three molecules of Br and NO_2 substituted chalcone derivatives namely (2*E*)-1-(4-bromophenyl)-3-[4(methylsulfanyl)phenyl]prop-2-en-1-one (4Br4MSP), (2*E*)-1-(3-bromophenyl)-3-[4-(methylsulfanyl) phenyl]prop-2-en-1-one (3Br4MSP) and (2*E*)-3[4(methylsulfanyl) phenyl]-1-(4-nitrophenyl)prop-2-en-1-one (4N4MSP). The NLO properties have been investigated by Z-scan technique using 2 ps laser pulses at 800 nm. The nonlinear refractive indices, nonlinear absorption coefficient, and the magnitude of third-order susceptibility have been determined. The values obtained are of the order of 10^{-7} cm^2/GW, 10^{-3} cm/GW and 10^{-14} esu respectively. The molecular second hyperpolarizability for the chalcone derivatives is of the order of 10^{-32} esu. The coupling factor, excited state cross section, ground state cross section etc. were determined. The optical limiting (OL) property was studied. The results suggest that the nonlinear properties investigated for present chalcones are comparable with some of the reported chalcone derivatives and can be desirable for NLO applications.

1. Introduction

The application of nonlinear optics in optoelectronic and photonic devices grasped the attention on nonlinear optical materials. Especially, nonlinear optical materials exhibiting strong two-photon absorption (TPA) are in great demand, due to their applications in three-dimensional fluorescence imaging and multi-photon microscopy, eye and sensor protection, frequency up conversion lasing, optical signal reshaping and stabilizing fast fluctuations of laser power [1–5]. There is a need to design and develop novel nonlinear materials with large molecular two-photon absorption cross-sections to meet the present demand. However, such attempts toward understanding the third-order NLO properties of organic crystals are very limited [6–8]. Organic materials are attractive because of their optical and electronic properties which can be tuned by structural modifications. Organic materials with large third-order nonlinear optical (NLO) properties will be the key elements for future photonic technologies [9–11]. The large and ultrafast NLO response has made organic molecules attractive candidates for high bandwidth applications. Chalcones belong to the class of low molecular charge transfer (CT) compounds that show strong nonlinear optical properties without longer wavelength absorption. These emerged as promising candidates for third order nonlinear optics because of their noticeable third order nonlinearity and good optical power limiting properties [12–15]. Since the chalcone backbone is an asymmetric transmitter, it strongly increases the molecular nonlinearity for the electron donating and withdrawing group substitutions. Generally in charge transfer compounds, large nonlinear electronic polarization arises due to the large dipole moment change from ground state to an excited state by optical radiation [16]. The other advantage of chalcone molecules is that it offers greater flexibility in adopting suitable design strategies [17,18], and thereby enhancing the nonlinear optical coefficients. In this paper we report our investigations on third ordered nonlinear optical properties of three chalcone molecules namely, (2*E*)-1-(4-bromophenyl)-3-[4-(methylsulfanyl)phenyl]prop-2-en-1-one (4Br4MSP), (2*E*)-1-(3-bromophenyl)-3-[4 (methylsulfanyl) phenyl]prop-2-en-1-one (3Br4MSP) and (2*E*)-3[4(methylsulfanyl) phenyl]-1-(4-nitrophenyl)prop-2-en-1-one (4N4MSP) by adopting the picosecond (ps) Z-scan technique. The ps studies are important to identify the electronic part of nonlinearity. Open aperture data provided the nonlinear absorption coefficients while the closed aperture data provided nonlinear refractive index. The contribution of solvent is also identified.

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2. Experimental procedure

The compounds were synthesized by following the standard procedure namely Clainsen–Schmidt condensation method [19–21]. This is a reaction of substituted acetophenone with substituted benzaldehyde in the presence of an alkali. Commercially available 4-bromoacetophenone, 3-bromoacetophenone, 4-nitroacetophenone was treated with 4-methylthiobenzaldehyde to get 4Br4MSP, 3Br4MSP, 4N4MSP respectively. The compounds were used without further purification. The synthesis of the compounds follows literature method. The resulting crude after synthesis reaction was collected by filtration, dried and purified by repeated crystallization from acetone. The structures of the compounds are shown in Fig. 1. The linear absorption spectra of the samples recorded at room temperature in dilute DMF solution (2×10^{-2} mol/L) using a spectrophotometer (SHIMADZU UV-1800) is shown in Fig. 2. The linear absorption spectrum of solutions shows that all the compounds are transparent at 800 nm. The linear refractive index of these samples dissolved in DMF was measured using an Abbe refractometer. The single beam Z-scan technique was employed to study the nonlinear optical properties of these compounds [22,23]. This technique is particularly useful when the nonlinear refraction is accompanied by nonlinear absorption and allows one to measure both sign and magnitude of the nonlinear refractive index. The transmittance of the nonlinear medium through a finite aperture in the far field as a function of the sample position is measured with respect to the focal plane of the focused Gaussian laser beam. Here the sample itself acts as a thin lens with varying focal length as it moves through the focus. When the intensity of the laser beam is sufficient to induce the nonlinearity in the sample, the laser beam either focuses or defocuses depending upon the nature of the nonlinearity of the material.

The experiment is performed by using Z-scan experimental setup with a ps amplifier [seeded by Micra laser oscillator (COHERENT)] generating pulses of duration ~2 ps at a repetition of 1 kHz. The experiment has been done at 800 nm wavelength only. The maximum average and peak powers are ~2 W and ~1 GW, respectively. The diameter of the beam from the amplifier is 4 mm. Using ND filters power has been reduced to avoid sample damage. Sample solutions are prepared using DMF as a solvent. 4N4MSP, 3Br4MSP are made of 10 mM and 4Br4MSP is prepared with a concentration of 15 mM. The 800 nm beam is focused using a 20 cm biconvex lens and allowed to pass through a 1 mm quartz cuvette that contained sample solution. The resulting beam waist radius at the focus was 25.44 μm that corresponds to the Rayleigh length of 2.54 mm. The sample thickness of 1 mm is lesser than Rayleigh length and therefore could be treated as thin medium. The Z-scan performed at laser pulse energy of 1 mW, which resulted in an on-axis peak irradiance of 49 GW/cm^2. All the sample measurements were done at room temperature. Sample in the cuvette moved from the extreme end of the translation stage to the other end in steps of 1 mm, and transmitted power is noted down for each position of the sample using a power sensor (COHERENT Power Max, measuring range: 1 μW–1 W). We also demonstrate the strong optical power limiting property of ps laser pulses at 800 nm based on the two photon absorption (TPA) process. In the open aperture (OA) scheme the aperture of the power sensor is fully opened and transmission is noted down where as in closed aperture (CA) scheme the aperture of the power sensor is partially closed and the transmittance is recorded. In both of the cases plots had been taken for position versus normalized transmittance using MATLAB and ORIGIN. From the open aperture analysis we can find out the nonlinear absorption coefficient/two photon absorption coefficient (β). From the closed aperture analysis we can calculate intensity dependent refractive index (n_2) of the sample.

3. Result and discussion

The nonlinear absorption in the materials arising from either direct multi-photon absorption, saturation of the single photon absorption, or dynamic free carrier absorption has strong effect on the measurements of nonlinear refraction using the Z-scan technique. When Z-scan with open aperture ($S = 1$) is performed, will be insensitive to nonlinear refraction (thin sample approximation). Such Z-scan traces with no aperture are expected to be symmetric with respective to the focus ($z = 0$) where they have a minimum transmittance (e.g. multi-photon absorption) or maximum transmittance (e.g. saturation of absorption). In fact the coefficient of nonlinear absorption can be easily calculated from such transmittance curves. The third order nonlinear susceptibility is a complex quantity

$$\chi^{(3)} = \chi_R^{(3)} + i\chi_I^{(3)} \tag{1}$$

where the imaginary part is related to the 2PA coefficient β through

$$\chi_I^{(3)} = \frac{n_0^2 \varepsilon_0 c^2}{\omega}\beta \tag{2}$$

and the real part is related to γ through

$$\chi_R^{(3)} = 2n_0^2 \varepsilon_0 c\gamma \tag{3}$$

In the case of 2PA the nonlinear absorption coefficient β is related to linear absorption coefficient by the relation $\alpha(I) = \alpha_0 + \beta I$. This yields the irradiance distribution and phase shift of the beam at the exit surface of the sample as $I_e(z,r,t) = (I(z,r,t)e^{-\alpha L})/(1 + q(z,r,t))$ and $\Delta\phi(z,r,t) = (k\gamma/\beta)\ln[1 + q(z,r,t)]$, where $q(z,r,t) = \beta I(z,r,t)L_{eff}$. In the case of open aperture Z-scan ($S = 1$), for a Gaussian beam, the

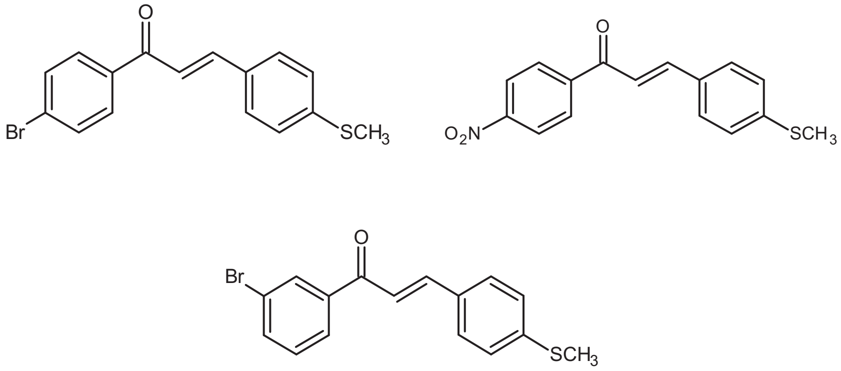

Fig. 1. Molecular structure of 4Br4MSP, 4N4MSP and 3Br4MSP.

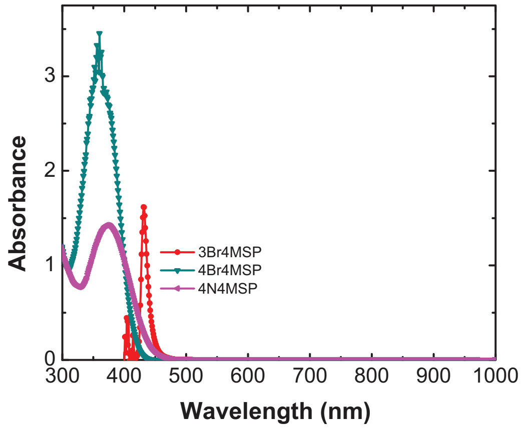

Fig. 2. Linear absorbance spectrum of chalcone derivatives.

transmittance can be given by the equation [22], $T(z) = \ln[1 + q_0(z)]/q_0(z)$ for $|q_0(z)| < 1$, where $q_0(z) = \beta I_0 L_{eff}/(1 + z^2/z_0^2)$.

For a cubic nonlinearity and a small phase it can be shown that the normalized transmittance in the case of closed aperture z-scan is

$$T(z) = 1 + \frac{4\Delta\phi_0 x}{(1+x^2)(9+x^2)} - \frac{2\Delta\phi_0(3+x^2)}{(1+x^2)(9+x^2)},$$

where $\Delta\phi_0 = \Delta T_{P-V}/0.406(1-S)^{0.25}$ for $|\Delta\phi_0| \leq \pi$ and $\Delta\phi_0 = (1/2)\beta I_0 L_{eff}$.

For a pure nonlinear refraction curve, obtained by division of closed aperture scan by open aperture scan, the normalized transmittance is shown to be

$$T(z) = 1 + \frac{4\Delta\phi_0 x}{(1+x^2)(9+x^2)}$$

Once $\Delta\phi_0$ is calculated, one can readily calculate γ using equation [24] $\gamma = \Delta\phi_0\lambda/2\pi L_{eff} I_0$, from which one can obtained n_2 through the conversion formula given by $n_2(\text{esu}) = (cn_0/40\pi)\gamma\,(\text{m}^2/\text{W})$.

The open aperture Z-scan curves are shown in Fig. 3. The experimental data was fitted with the theoretical formula for two photon absorption. The presence of valley in normalized transmittance in open aperture scans indicates strong reverse saturation absorption (RSA) at peak intensities. The closed aperture measurements were carried at the input energy of 1 mW. The nonlinear refraction (NLR) curves of the compounds are shown in Fig. 4. The valley followed by peak in the normalized transmittance data clearly suggests that the sample possesses positive type of nonlinearity and self-focusing behavior. In order to identify the contribution from the solvent DMF, the Z-scan was performed on it and observed same sign and a higher value of n_2 ($n_2 = 3 \times 10^{-16}$ cm^2/W) indicating that the solute had an opposite sign of nonlinearity (Fig. 5). The magnitudes of n_2 are estimated to be $\sim 10^{-16}$ cm^2/W. The slightly deviation for the fitted line and the experimental data for closed aperture fit of 4N4MSP may be due to thermal contribution to the nonlinearity, higher order nonlinear effect or nonlinear scattering [15]. The experimentally determined values of β_{eff}, n_2, Re χ^3 and Im χ^3 are given in the Table 1. The values calculated in Table 1 include the solvent contribution. The calculated value of $\chi^3 = \sqrt{(\text{Re}\,\chi^{(3)})^2 + (\text{Im}\,\chi^{(3)})^2}$ (using Eqs. (2) and (3)) for the samples is of the order of 10^{-14} esu and is influenced by different donor or acceptor group substitutions and their position on the aromatic ring in the molecular structure of chalcone [24]. The value of n_2 is found to be of the order of 10^{-14} esu.

Among the compounds studied, 4N4MSP shows high χ^3 value compared with 4Br4MSP. The Br acts as an electron donating group, whereas SCH_3 is also an electron donor group. 4Br4MSP can be considered as effective D–A–D (donor–acceptor–donor) type of molecule. In the case of 4N4MSP, the nitro group present is a strong acceptor group and a molecule is A–A–D type. In the case of D–A–D type of molecules charge transfer takes place through donor to acceptor and in A–A–D type through acceptor end to donor end (between two end groups of the molecules). χ^3 value found to increase with the presence of strong acceptor or donor groups. 3Br4MSP shows less value of χ^3 when compared with other two

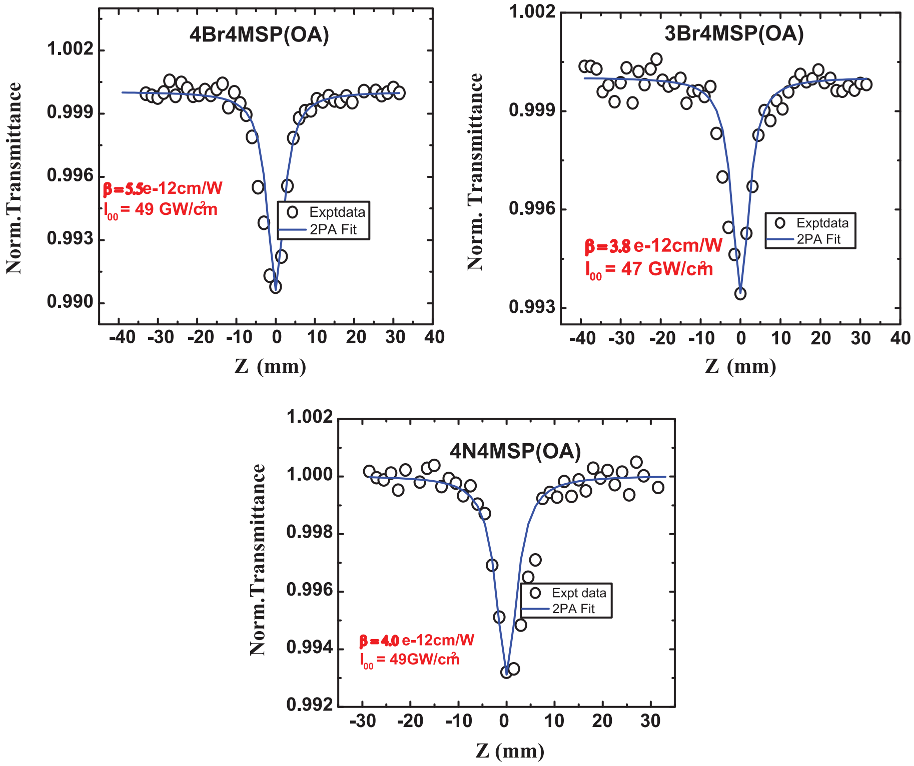

Fig. 3. Open Z-scan data of chalcone solutions of DMF. The open aperture Z-scan data represented by open circle and solid line is the fit to the experimental data.

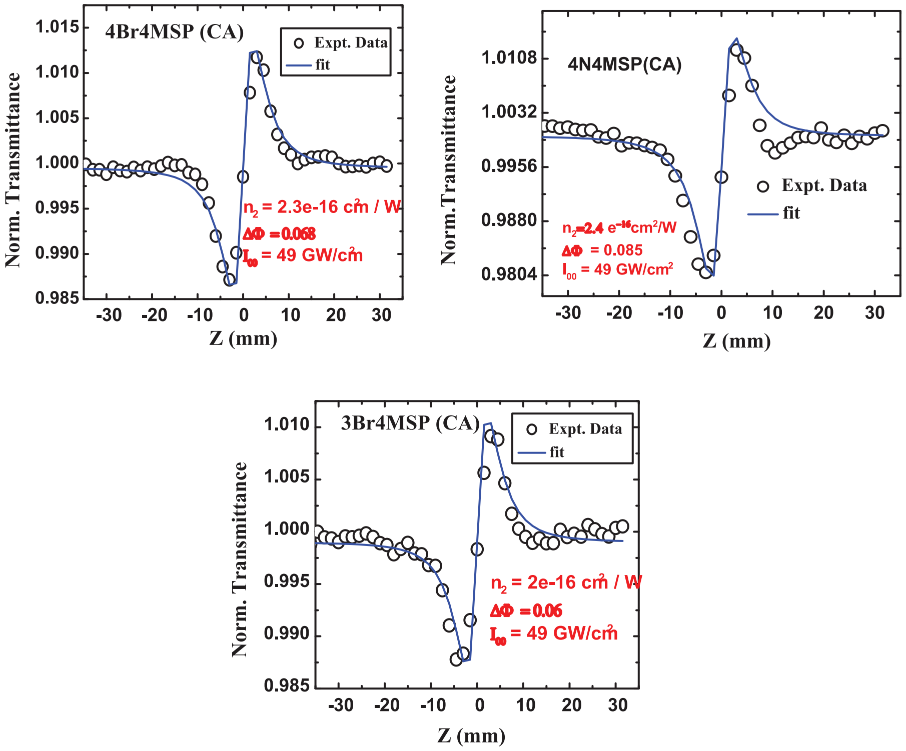

Fig. 4. Closed aperture Z-scan data of chalcone solutions of DMF. The closed aperture Z-scan data represented by open circle and solid line is the fit to the experimental data.

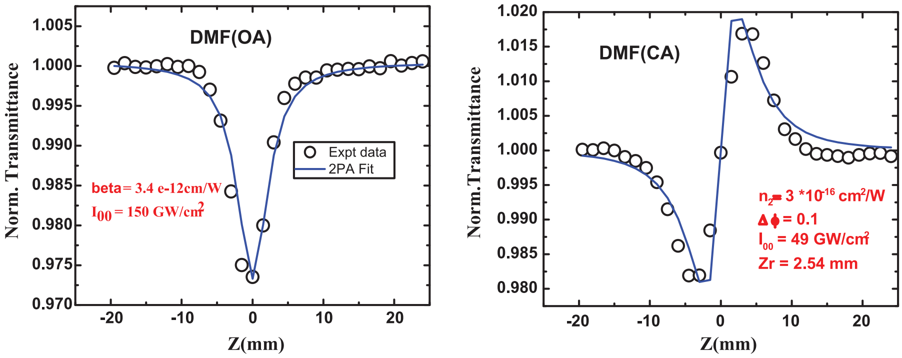

Fig. 5. Open and closed aperture Z-scan data of pure DMF solution.

Table 1
Third order nonlinear parameters of 4-methylsulfanyl chalcone derivatives.

Sample	n_0	n_2 ($\times10^{-7}$ cm^2/GW)	n_2 ($\times10^{-14}$ esu)	β_{eff} ($\times10^{-5}$ cm/GW)	Re$\lvert\chi^3\rvert$ ($\times10^{-22}$ m^2/V^2)	Im$\lvert\chi^3\rvert$ ($\times10^{-22}$ m^2/V^2)	$\lvert\chi^3\rvert$ ($\times10^{-19}$ m^2/V^2)	$\lvert\chi^3\rvert$ ($\times10^{-14}$ esu)
4Br4MSP	1.363	2.30	7.49	5.5	2.27	0.046	2.30	1.65
3Br4MSP	1.365	2.00	6.52	3.8	1.98	0.047	1.99	1.43
4N4MSP	1.360	2.40	7.79	4.0	2.36	0.097	2.37	1.70

molecules. Here in this molecules Br is present at the *meta* position on the aromatic ring and the length of the charge transfer axis is less compared to other two molecules. It was observed that as the length of the charge transfer axis contributes to nonlinearity [25]. The dihedral angle between aromatic rings is 47.82°, 50.03° respectively for 4Br4MSP and 3Br4MSP. The large dihedral angle of 3Br4MSP causes less efficient charge transfer through its backbone than compared with 4Br4MSP and show less nonlinearity. Third-order susceptibility values are of same order when compared with some chalcone derivatives reported earlier [15].

The coupling factor ρ, the ratio of imaginary part to real part of third-order nonlinear susceptibility can be measured as [24], $\rho = \mathrm{Im}\,\chi^{(3)}/\mathrm{Re}\,\chi^{(3)}$. The observed values of coupling factor for the samples 4Br4MSP, 3Br4MAP and 4N4MSP are 0.208, 0.121, and 0.106 respectively. The nonlinear induced polarization per molecule is described by the microscopic susceptibilities known as the hyperpolarizabilities. For third-order effects the corresponding hyperpolarizability γ_h (second order hyperpolarizability) is related to the third order susceptibility $\chi^{(3)}$ by the equation [26]

$$\gamma_h = \frac{\chi^{(3)}}{[(1/3)(n_0^2+2)]^4 N}$$

Table 2
Linear and non-linear optical parameters of chalcones.

Sample	α (mm^{-1})	γ_h ($\times 10^{-32}$ esu)	σ_2($\times 10^{-46}$ cm^4 s/photon)	σ_{ex} ($\times 10^{-19}$ cm^2)	σ_g ($\times 10^{-19}$ cm^2)
4Br4MSP	0.116	0.070	1.51	11.67	1.29
3Br4MSP	0.163	0.100	1.56	5.78	2.69
4N4MSP	0.151	0.104	1.65	6.55	2.51

where N is the density of molecules in the unit of molecules per cm^3 and n_0 is the linear refractive index of the medium. The value of γ_h obtained are tabulated in Table 2. The γ_h of 4N4MSP and 3Br4MSP is one order less compared with the reported values of polymer doped chalcones [14,27] and have equal order with the values reported for chalcones by Ravindra et al. [15]. But the value of γ_h 4Br4MSP is found to be one order less than the literature value [15].

The two photon absorption (TPA) cross section, σ_2 (in units of cm^4 s/photon), which defines the transition rate for TPA was calculated for all the samples using the equation [24] $\beta_{eff} = \sigma_2 N_A d \times 10^{-3}/h\nu$, where N_A is the Avogadro number, d is the concentration of the samples in mol/L and $h\nu$ is the energy of an incident photon (in J). The measured σ_2 values are in the order of 10^{-46} cm^4 s/photon (Table 2). ESA data is of interest because on one hand it provides fundamental information about the nature of the lying excited states often inaccessible from the ground state, and on the other hand, it can provide important information to assess the lasing potential of the medium [28].

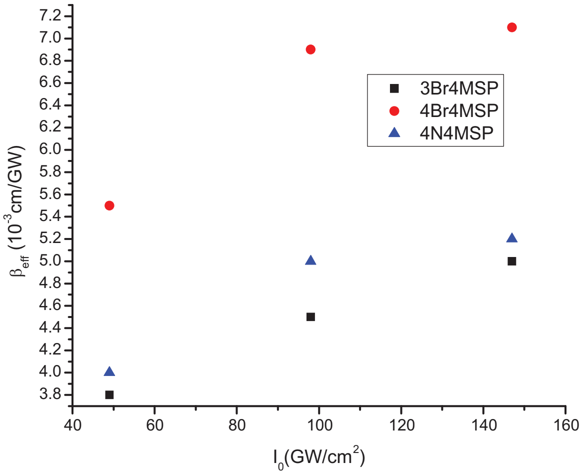

Fig. 6. A plot of β_{eff} vs. I_0 for chalcone derivatives.

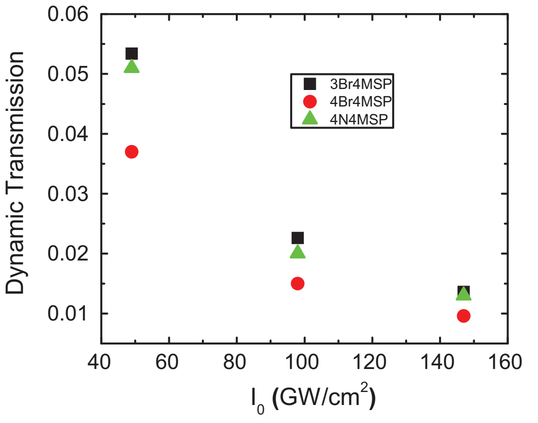

Fig. 7. A plot of dynamic transmission vs. intensity for chalcone derivatives.

The TPA cross section depends on type of functional group and its position on the aromatic ring of the chalcone molecule [24]. The excited state cross section σ_{ex} (Table 2) for all the samples can be determined from the procedure described in the literature [29]. The ground state absorption cross section σ_g (Table 2) can be calculated from the equation $\alpha = \sigma_g N_A C$, where N_A is the Avogadro number, C is the concentration in mol/cm^3 and α is the linear absorption coefficient. The value of σ_{ex} was found to be larger than the value of σ_g, which is in agreement with the condition for observing reverse saturable absorption [30].

The knowledge of the on-axis irradiance dependence of the nonlinear absorption (NLA) coefficient gives the information about the mechanism of the nonlinear absorption. If the mechanism belongs to simple two photon absorption (TPA), then β_{eff} should be a constant that is independent of the on-axis peak irradiance I_0. If the mechanism is due to direct three-photon absorption then β_{eff} should be a linearly increasing function of I_0 and the intercept at the vertical axis must be zero [31]. We used three different input intensities I_0 = 49 GW/cm^2, I_0 = 98 GW/cm^2, I_0 = 147 GW/cm^2 to measure β_{eff}. It is observed that β_{eff} increases with increase in I_0 (Fig. 6) even with the excited-state absorption being greater than that of the ground state and the intercept on the vertical axis is non zero, this suggests that a higher order effect, such as excited state absorption (ESA) via two photon absorption, could be contributing to the nonlinear absorption (NLA).

Optical limiting is a phenomena observed when the transmission of a medium decreases with increasing input laser intensity (or fluence). 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 de-focusing along with MPA [32]. The present chalcone samples have negligible linear absorption at the operating laser wavelength 800 nm. The relation for intensity dependant nonlinear absorption coefficient is given by [14]

$$\frac{dI}{dz} = -\beta I^2 \qquad (4)$$

where β is the nonlinear absorption coefficient. The solution of Eq. (4) may be written as $I(z) = I_0/(1+\beta z I_0)$, where I_0 is the input intensity. Hence dynamic transmissivity can be given by $T(I_0, z) = I(z)/I_0 = 1/(1+\beta z I_0)$. We plotted the graph of transmission versus input peak laser intensity, is shown in Fig. 7. An effective optical limiter will have low limiting threshold, high optical damage threshold and stability, fast response time, high linear transmittance throughout the sensor bandwidth, optical clarity and robustness. In the case of present chalcone samples, it is observed that sample exhibits strong two-photon absorption and therefore is responsible for the limiting reported here.

4. Conclusions

The third order nonlinear optical properties of donor and acceptor substituted chalcones was investigated at 800 nm using picosecond laser pulses by Z-scan technique. The dependence of nonlinear responses of these sample molecules on substituent groups is discussed. The presence of strong donor or acceptor functional group enhanced the third-order susceptibility χ^3 and second order hyperpolarizability γ_h which can be comparable with some of reported chalcone molecules. All the three samples show pleasing optical limiting behavior at 800 nm. The present results show that the samples exhibit satisfactory optical responses in the picosecond domain thus can be considered for designing suitable optical devices for eye and sensor protection, optical limiting and for electronic applications.

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