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Characterizing the passive modelocker for an ultrafast laser

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Dedicated to Prof Kehar Singh on the occasion of his 73rd birthday

An economic, solid state device for efficient modelocking the ultrafast lasers is the contemporary area of research. In this paper we have studied the nonlinear optical properties of a standard dye IR26 by using the Z-scan technique at 1064 nm as a function of its concentration. A prominent contribution of nonlinear absorption is observed in the Z-scan profiles. The commercial laser has been checked for its modelocking efficiencies at various concentrations to provide the typical values for a standard modelocker.

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1 Introduction

Pupil filters have been widely used to improve the optical response of an optical system. Non-uniform amplitude-only filters produce effects like apodization or superresolution on the Point Spread Function (PSF) [1]. These types of filters have been applied in very different fields. In other cases, apodizers have been proposed to reduce the effect of aberrations [2-3]. Annular pupils have been widely used to produce lateral superresolution [4]. The use of annular color filters to control the polychromatic response of an optical filter was also proposed [5]. Different superresolution pupils have also been sought through amplitude-only filters in fields like confocal microscopy [6], in some cases combined with polarized light [7] and in microlithography [8-9].

Heptamethine cyanine dyes are important class of chromophores with absorption and fluorescence spectra in visible and near infrared regions [1]. Their high photostability combined with the high molar extinction coefficient leads to their application as modelocking in lasers [2]. A series of distinguished multifunctional chromophores, cyanine dyes and their J and H-aggregates [3] have recently evoked interest in the areas of optoelectronics and nonlinear optics. One of the standard heptamethine cyanine dye IR26 has been used for passive modelocking in commercial lasers [4]. Its nonlinear optical properties have been studied recently by us [5].

Z-scan technique [6] is used to measure the nonlinear refractive (NLR) and nonlinear absorptive (NLA) parameters of a material. 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).

The semiconductor saturable absorber mirror (SESAM) technology [7] has been used for modelocking over the last decade. However, this is an expensive technique. In this direction we have been working towards economic alternates such as a new class of highly efficient Weinreb amide based dyes [8], gold nanoparticle embedded hetro structures [9] and nanoparticle embedded graphenes [10]. In any case, even if the material exhibits the saturable absorption, it is important to characterize it with its suitable optical density, optical thickness and the extinction coefficient at the wavelength of interest. In view of the above,

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in this paper, we have studied a standard saturable absorber to provide these parameters for any material for modelcoking applications.

2 Experimental

The absorption spectrum was recorded using a UV-visible spectrometer (JASCO, V-570) in quartz cell of 1 mm thickness. The experimental arrangement for the open aperture Z-scan is given in Fig 1. The fundamental (1064 nm) wavelength of a picosecond Nd: YAG laser (Continuum Model YG601, 30 ps, 10 Hz) were used for the Z-scan experiments. The dye IR26 (Lambdachrome) and the solvent 1, 2-dichloroethane (Aldrich) were used as received. The transmitted energy as a function of the sample position at the far field was measured with the help of the photodiode (Becker and Hickle, PDI-400) after focusing with a double convex lens of focal length of 50 mm.

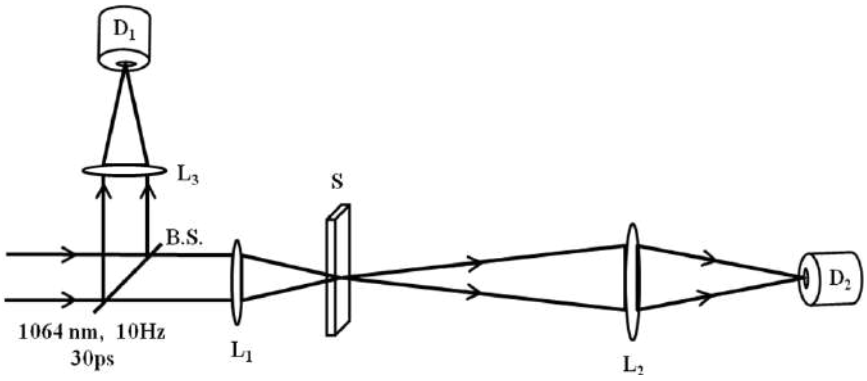

Fig 1. Open aperture Z-scan experiment. B.S. is the beam splitter, L1, L2 and L3 are double convex lenses, S is sample, D1 and D2 are reference and signal photo diodes, respectively.

3 Theory

The polarization of materials at the strength of interatomic filed (~10^5 V/m) can be expressed as

$$P_i(t) = \varepsilon_0 \left[\sum_j \chi_{ij} E_j + \sum_{jk} \chi^{(2)}_{ijk} E_j E_k + \sum_{jkl} \chi^{(3)}_{ijkl} E_j E_k E_l + ... \right] \quad (1)$$

Here, $P_i(t)$ is the i^{th} component of polarization, ε_0 is the permittivity of free space, χ_{ij} is the electric susceptibility, E represents the applied electric field, and i, j, l are the Cartesian coordinates. Here χ_{ij} is a dimensionless quantity. $\chi^{(2)}_{ijk}$, and $\chi^{(3)}_{ijkl}$ are the second and third order nonlinear susceptibilities, respectively. $\chi^{(2)}_{ijk}$ has the units of m/v while that of $\chi^{(3)}_{ijkl}$ is m^2/v^2. Generally, for centro symmetric materials the values of even order nonlinear susceptibilities (e.g. $\chi^{(2)}$, $\chi^{(4)}$ etc) are zero. The optical properties of a medium become dependent on the incident light at higher intensities.

In the presence of high irradiance (I), the total refractive index (n (I)) and the total absorption coefficient (α (I)) of a Kerr like sample not only depend on linear indices (n_0 and α_0, respectively) but also become functions of nonlinear indices (η_2 (m^2/W) and β (m/W), respectively) as follows

$$\alpha(I) = \alpha_0 + \beta I \quad (2)$$

$$n(I) = n_0 + \eta_2 I \quad (3)$$

Here β and η_2 are cubic nonlinear absorption and refractive indices,respectively. These are related to the intensity and phase modulation of the Gaussian beam, respectively as.

$$\beta = \frac{2\Delta\psi}{I_0 L_{\text{eff}}} \tag{4}$$

$$\eta_2 = \frac{\Delta\phi}{kI_0 L_{\text{eff}}} \tag{5}$$

Here $L_{\text{eff}} = [1 - \exp(-\alpha_0 L)]/\alpha_0$ is the effective length of the sample and $\Delta\phi$ and $\Delta\Psi$ are the on-axis phase shifts due to refractive nonlinearity and absorptive nonlinearity, respectively. The phase and intensity modulated normalized transmittance in the closed aperture Z-scan experiment for the cubic nonlinearity is given by [11].

$$T(z) = 1 + \frac{4x}{(x^2+9)(x^2+1)}\Delta\phi - \frac{2(x^2+3)}{(x^2+9)(x^2+1)}\Delta\psi \tag{6}$$

Here $x = z/z_0$, in which z_0 ($= \pi\omega_0^2/\lambda$) is the Rayleigh range with ω_0 as the beam waist at the focus of the pump wavelength (λ). In open aperture Z-scan experiment, the transmitted signal is insensitive to the beam distortion due to the refractive part and has only contribution from the material absorption [12].

$$T(z) = 1 - \frac{1}{2(x^2+1)}\Delta\psi \tag{7}$$

The real and imaginary part of the third order nonlinear susceptibility ($\chi^{(3)}$) can be expressed as

$$\chi_R^{(3)}(\text{esu}) = \frac{cn_0^2}{120\pi^2}\eta_2 \ (\text{m}^2/\text{W}) \tag{8}$$

$$\chi_I^{(3)}(\text{esu}) = \frac{c^2 n_0^2}{240\pi^2\omega}\beta(\text{m/W}) \tag{9}$$

The absolute $\chi^{(3)}$ can be obtained by

$$|\chi^{(3)}|^2 = |\chi_R^{(3)}|^2 + |\chi_I^{(3)}|^2 \tag{10}$$

The incident field induced change in the absorption coefficient (third order nonlinear response) is responsible for the passive modelocking.

4 Results and discussion

4.1. Z-scan profiles

Absorption spectra of dye IR 26 in a solvent are shown in Fig 2 (A). The dye has strong absorption at the fundamental wavelength region of the Nd:YAG laser. This makes it suitable for the modelocking of this laser. The optical density of the dye increases without appearance of any new band suggesting that there is no dimerisation in the ground state. Figure 2 (B) shows the OA Z-scan profiles recorded at 1064 nm for two dye concentrations at a fixed energy of 1.6 μJ. The profile shows the peak at the focus indicating the effect of saturable absorption (SA). We have not observed any dip at the center [5] due to the removal of the artifacts in the alignment. The experimental data is fitted with the Eq (7). From this profile, the imaginary part of the third order nonlinear susceptibility ($\chi_R^{(3)}$) was determined to be -4.1×10^{-11} (esu) (-5.7×10^{-19} $\underline{\text{m}}^2/\underline{\text{V}}^2$).

4.2. Application in passive mode-locking

Mode-locking can be achieved either by an electronic device (active mode-locking), or a passive device using a material with SA property (passive mode-locking) or with both devices (active-passive hybrid

mode-locking). In view of the demanding industrial application, we characterize the dye for its suitable parameters for passive mode-locking.

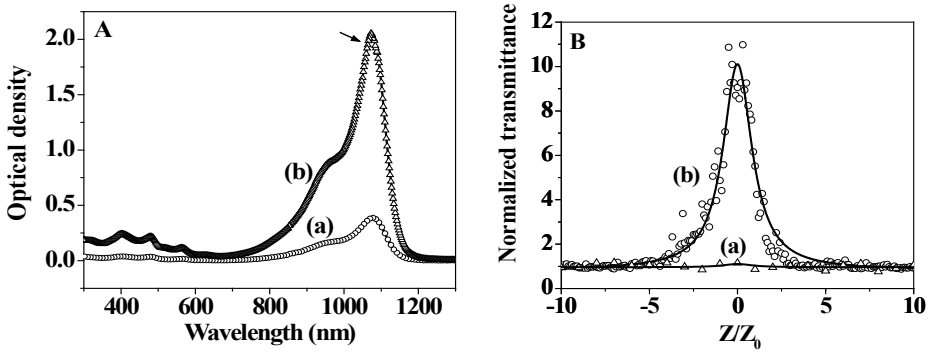

Fig 2. The absorption spectrum of the dye IR26 in 1, 2, dichloroethane (Panel A). The concentration of the dye (b) is higher than (a) needed for modelocking the laser. Arrow indicates the pump wavelength in Z-scan experiment. Panel B gives the OA Z-scan profile of the dye IR26 at 1064 nm at a constant energy of 1.6 μJ.

4.2.1. Ultrafast time response

One of the major requirements for the efficient passive mode-locking is the fast decay/recovery time of the SA material. The time-resoponse of the optical nonlinearity must be fast enough for generation of the short pulses. To have the estimation of the decay time of these samples, the time-resolved decay of one of the sample was measured by using the box geometry of the laser induced transient grating (LITG) technique [13]. To calibrate the measurement system, first the decay profile of a standard dye R6G was measured. For IR 26, the relaxation time of ~ 20 ps was obtained indicating that the dye can be used as a fast SA material in ps lasers.

4.2.2. Figures of merit values for all-optical switching applications

If a nonlinear material is to be used for photonic switching applications, it should satisfy the Stegeman relations [14]. For SA materials, the effect of linear absorption must be weak compared to the effect of nonlinear effect. This condition quantifies in terms of single photon figure of merit value (W) given as,

$$W = \frac{\Delta n}{\alpha_0 \lambda} > 1 \tag{11}$$

where Δn is induced refractive index change ($= \eta_2 I_0$). The estimated value of W for IR 26 is 1.7 indicating that it can be used for the optical switching devices.

4.2.3. Passive mode-locking

IR26 was taken in solution in a dye cell kept at one end of the cavity mirrors in the commercial laser which is actively modelocked. Figure 3 shows the direct oscilloscope traces of the pulse generation monitored from the laser cavity. It can be seen that in free running mode (Panel A) there are six steps in the trace consisting of various modes of the laser within a single flash of the pump. The number of steps decreases on increasing the concentration of the dye (increment level of 5 μM) due to suppression of the lower intensity modes. We can see that in Panel D, there is only one step suggesting that there is only one strong set of modes within the time duration of the flash lamp. By adding further concentrations of IR26

dye (Panel E), we can further reduce the wings of the pulse train. Panel F shows the singe pulse extracted from the cavity after cavity dumping and after passing through the amplifier.

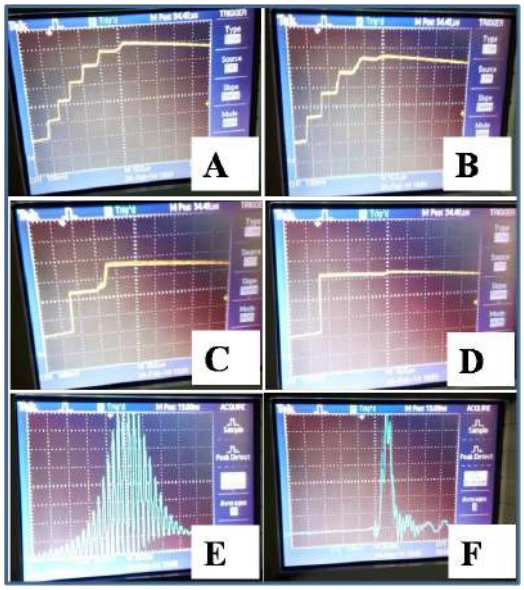

Fig 3. Panels A-D shows the direct photographs from the oscilloscope monitoring the intracavity pulses as a function of concentration of IR 26 dye (at 1 MΩ input impedance of the oscilloscope). Panel E is the modelocked pulse train at 80 MHz before cavity dumping (at 50 Ω impedance of the oscilloscope). Panel F is the single pulse output amplified after cavity dumping and passing through the amplifier Nd:YAG rod. Time scale of Panels A to B is in 100 μs, while that in Panels E and F is 25 ns. The detector response is slower than the actual pulse (~30 ps).

5 Conclusions

We have measured the nonlinear optical response of the IR26 using Z-scan technique at two different concentrations. At 1064 nm, at relatively lower pulse energies the optical nonlinearity in the Z-scan profiles are not observed (at the concentrations required for the passively modelocking the laser). However, a 5 fold increase in the concentration results in a prominent effect of SA in the sample. The passive modelocking sequence of a Nd:YAG laser has been shown as a function of the dye concentration due to the SA nature of the optical nonlinearity. It is, therefore, concluded that the required thickness or the optical density of the materials must be smaller than those required for the modelocking applications.

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