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$\chi^{(3)}$ Measurement of Ferronematic Liquid Crystal Using a Single Beam Method

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Abstract

The suspension of magnetic particles in nematic liquid crystals (NLCs), usually called ferronematic (FN). The FN studied in this work are E7 doped with small compositional percentage of Fe_3O_4 nps (5% W/W). Synthesis of the spherical Fe_3O_4 nps was based on efficient chemical co-precipitation technique. Two orientations of molecules of a FN (homogeneous and homeotropic) were used for studying third order nonlinear susceptibilities, $\chi^{(3)}$ measurement by a single beam z-scan technique. The absolute values of $\chi^{(3)}$ were in the order of 10^{-5} esufor homogeneous and homeotropic alignment. The experimental results showed that samples possess a self-defocusing optical nonlinearity and two photon absorption (TPA) effect for homogeneous cells while self-focusing and saturable absorption (SA) effects in homeotropic cells.

Keywords: Ferronematic; Nanoparticles; Third order nonlinear susceptibility; Homogeneous; Homeotropic.

Introduction

In recent years materials with excellent third order nonlinear optical (NLO) properties have gained much attention due to their potential applications such as optical power limiting, optical switching, data processing, ultra-fast optical communications, 3-D optical memory devices, optoelectronic and photonic devices [1-3]. Composite materials consisting of liquid crystals doped with nps have indeed attracted much scientific and technological interest mainly because the incorporation of nanomaterials enhances the optical properties of the liquid crystal itself [4].

Dielectric Polarization density, P (in terms of the electrical field, E) in nonlinear medium is:

$$P=\varepsilon_0[\chi^{(1)}E+\chi^{(2)}E^2+\chi^{(3)}E^3+...] \qquad (1)$$

Here, the coefficients $\chi^{(n)}$ are the n-th order susceptibilities of the medium and the presence of such a term is generally referred to as an n-th order nonlinearity [5, 6].

The z-scan method provides a sensitive and straight forward method for the determination of the sign and the values of the real and imaginary parts of $\chi^{(3)}$, respectively, proportional to the nonlinear refractive index (NLR), n_2 and the nonlinear absorption coefficient(NLA), β [7,8].

In this paper we present the results of third order nonlinear optical properties of FN measured by with the single beam z-scan technique with CW Nd: YAG laser at 532 nm. The values of the NLR index, n_2, NLA coefficient, β and third order nonlinear susceptibilities, $\chi^{(3)}$ are calculated.

Materials and Methods

Preparation of FN

Synthesis of Fe_3O_4 *nps*

To obtain Fe_3O_4 precipitate, aqueous solution of $FeCl_2$ $.4H_2O$ and $FeCl_3$ $.6H_2O$ were dissolved in deionized water by vigorous stirring. The sodium hydroxide solution was prepared by dissolving NaOH into de-ionized water. Then, under ultrasonic agitation and reaction temperatures 70 °C, NaOH was added to the prepared solutions drop by drop [9]. To purify prepared Fe_3O_4 nps and separating NaCl, the samples were washed repeatedly with de-ionized water. Because of attractive Vander Waals forces and additional electrostatic forces, these small nps would finally aggregate together and fall out. So, with the intention of preclude agglomeration and segregation, the particles were coated with a monomolecular film of Dodecyl-trimethyl ammonium bromide (DTAB) as surfactant [10]. The structure and the morphological investigation of the studied sample were taken by X-ray diffraction (XRD) and scanning electron microscopy (SEM), respectively. Then the nps were incorporated into a thermotropicnematic liquid crystal E7 to form ferronematics.

Liquid crystal cell preparation

Aligned monodomain cells were made up of two glass slides separated by Mylar sheets having 28 µm thickness. Before constructing of the cells, glass substrates were dip coated with polyvinyl alcohol (PVA) and lecithin for homogeneous alignment and homeotropic alignment; respectively. FN samples were filled into empty cells at a temperature higher than the isotropic temperature of the NLC via capillary action technique [11,12]. The compositional percentage of Fe_3O_4 nps in NLC mixtures is 5% W/W. They were used to determine the $\chi^{(3)}$ measurement of FN.

Third order susceptibilitymeasurement

The laser source has a Gaussian beam profile so the electric field variation is:

$$E(z,r,t)=E_0(t)\frac{\omega_0}{\omega(z)}\exp\left(-\frac{r^2}{\omega^2(z)}-\frac{ikr^2}{2R(z)}\right)\exp(-i\varphi(z,t)) \quad (2)$$

Where $\omega^2(z)=\omega_0{}^2(z)(1+z^2/z_0^2)$ is the spot size in z, $R(z)=1+z_0^2/z^2$ is the radius curvature, $z_0=\pi\omega_0^2/\lambda$ is the Rayleigh length of the beam and ω_0 is the beam radius at the focal point [13].

The $\chi^{(3)}$ measurements were performed using CW Ne:Yag laser at 532 nm. The beam waist radius of the focused Gaussian beam was about 34 µm (determined from edge scan method). In thez-scan measurements, the sample was held in a 28 µm cell under the on-axis peak intensity $I_0$of 550.7 W/cm^2. z_0 is the Rayleigh range given by $z_0=\pi\omega_0^2/\lambda$ [14] and was found to be 6.82 mm. The sample thickness was less than Rayleigh length and therefore could be treated as thin medium [3, 15]. In the experiment, the laser beam propagation direction is considered as the z-axis. The Gaussian laser beam is focused using a convex lens of focal length 5 cm, into the sample. In z-scan technique wih (close aperture) and without (open aperture) aperture, the samples are scanned on the z-axis, and the corresponding transmissions are measured. All the sample measurements were done at room temperature.

The third order nonlinear susceptibility is a complex quantity [3]

$$\chi^{(3)}=\mathrm{Re}(\chi^{(3)})+i\,\mathrm{Im}(\chi^{(3)}) \qquad (3)$$

$\mathrm{Re}(\chi^{(3)})$ *Measurement*

The real part of third order nonlinear optical susceptibility is obtained according to the following relations [15-17],

$$\mathrm{Re}(\chi^{(3)})(esu)=10^{-4}\varepsilon_0c^2n_0^2n_2(cm^2/W)/\pi \qquad (4)$$

Where ε_0 is the vacuum permittivity and c is the light velocity in vacuum and n_2 is the NLR index. The linear refractive index n_0 is obtained to be 1.7.

The difference between the normalized peak transmittance and valley transmittance, ΔT_{p-v} in the pure close aperture curve is used to calculate the NLR index, n_2 of the compounds through the equation [18]:

$$\Delta T_{p-v}=0.406\,(1-S)^{0.25}(2\pi/\lambda)n_2I_0L_{eff} \quad (5)$$

Where S is the aperture's linear transmittance(S= 0.32), $L_{eff}=(1-e^{-\alpha L})/\alpha$ is the effective thickness of the sample, α is the linear (low intensity) absorption coefficient, L is the thickness of samples, $I_0=2P_{in}/\pi\omega_0^2$ is the incident intensity at focal point, P_{in} is the laser power that is 10 mW in this work and λ is the wavelength of laser light.

$\mathrm{Im}(\chi^{(3)})$ *Measurement*

The imaginary part of third order nonlinear optical susceptibility is related to the NLA coefficient β, using the relation, [15-17]

$$\mathrm{Im}(\chi^{(3)})(esu)=10^{-2}\varepsilon_0c^2n_0^2\lambda\beta(cm/W)/4\pi^2 \qquad (6)$$

NLA coefficient was measured applying the open aperture z-scan technique. In this case (S = 1), for a Gaussian beam, the transmittance can be given by the equation,

$$T_{norm}(z)=Ln(1+q_0(z,t))/q_0(z,t) \qquad (7)$$

Where $q_0(z,t)=\beta I_0L_{eff}/(1+z^2/z_0^2)$, $z_0=k\omega_0^2/2$ is the diffraction length of the beam, z is the sample position and k is the wave vector [19-21].

Results

Characterization of Fe_3O_4 nps

The X-ray diffraction patterns of the synthesized nanoparticles are shown in Fig.1.

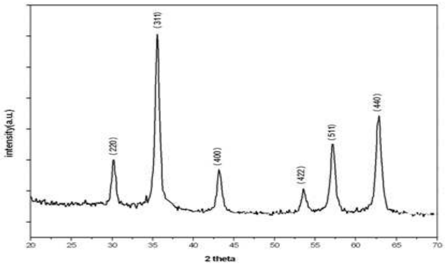

Fig.1: XRD pattern of synthesized Fe_3O_4 nps.

The sharpness of XRD reflections clearly shows highly crystalline structure and all the reflections in the patterns correspond to the cubic structure. The average size of the individual magnetite nanoparticles was calculated to be 20 nm using the Sherrer's equation [13].

The particles size and morphology are observed by SEM image, presented in Fig.2. It can be seen that the synthesized Fe_3O_4 consist of spherical particles with an average size 34 nm.

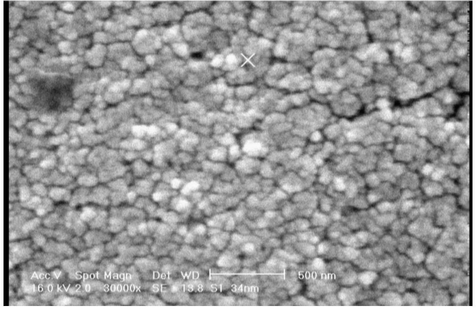

Fig.2: SEM image of synthesized Fe_3O_4 nps.

Nonlinear refraction

From the closed aperture analysis we can calculate NLR index (n_2) of the sample.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, it either converges (self-focusing) or diverges (self-defocusing) the beam, depending on the nature of the nonlinearity of the material [3, 15]. The NLO refractive effects were assessed by dividing the normalized z-scan data obtained under closed aperture configuration by the normalized z-scan data obtained under the open aperture configuration [22, 15]. Fig. 3a and b display the NLO refractive properties of FN for homogeneous alignment and homeotropic alignment; respectively.

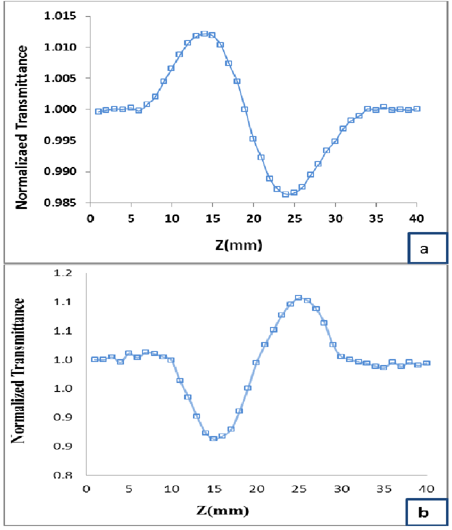

Fig 3.Normalized pure nonlinear refraction curve for FN in a) homogeneous alignment b)homeotropic alignment.

For homogeneous alignment configuration, the transmittance change has a peak- valley shape that is characteristic of a negative nonlinearity (self-defocusing effect). The valley-peak characteristic suggests that the NLR index is positive (self-focusing effect) for homeotropic alignment.

The NLR indices are obtained by fitting the experimental data using Eq. (4). The real part of third order nonlinear optical susceptibility, $\mathrm{Re}(\chi^{(3)})$ can be determined using the Eq. (5). The determined values of n_2 and $\mathrm{Re}(\chi^{(3)})$ for Fe_3O_4 nps in NLCs for both alignment are given in table 1.

Nonlinear absorption

In order to extract the information on NLA coefficient, the sample is moved through the focal point and the nonlinear transmission was measured as a function of sample position without an aperture (open aperture z-scan). Fig. 4a and b shows open aperture curves of FN in homogeneous alignment and homeotropic alignment, respectively.

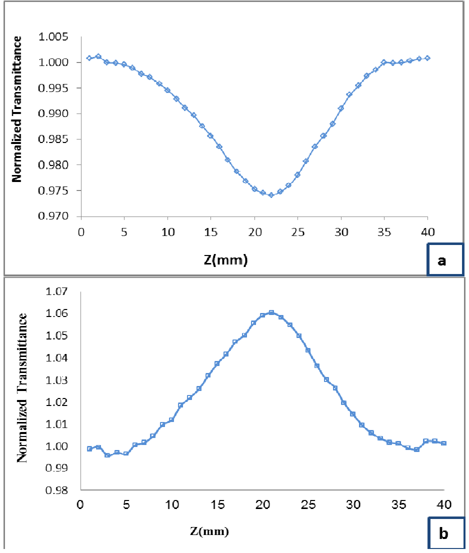

Fig. 4.Z-scan data for FN obtained under an open aperture configuration in a) homogeneous alignment b)homeotropic alignment.

The signature of the scans implies the intensity dependent variation of absorption. If the nonlinear transmissivity change is due to the pure two photon absorption (TPA) process, where it has a minimum transmission, showing intensity dependent absorption effect [15].Analyzing from open aperture data, the NLA may be mainly attributed to TPA effect for homogeneous alignment and saturable absorption (SA) for homeotropic alignment.

According to Eq. (7) the values of β correspond to $\mathrm{Im}(\chi^{(3)})$. By determining the best-fit curves for the experimental data, the nonlinear parameters β are found. The calculated β and $\mathrm{Im}(\chi^{(3)})$ values of FN are tabulated in table 1 for both alignments.

The absolute value of $\chi^{(3)}$ was calculated from below Eq. [16] and listed in table 1.

$$|\chi^{(3)}| = [(\mathrm{Re}(\chi^{(3)}))^2 + (\mathrm{Im}(\chi^{(3)}))^2]^{1/2} \qquad (8)$$

However, as described above, all the values obtained for homeotropic alignment including n_2, β, and $\chi^{(3)}$ are larger than those of homogeneous alignment. Such large values for homeotropic alignment are due to positive birefringence (i.e. $\Delta n \sim 0.13$) of E7) [23] that means the extraordinary refractive index is larger than ordinary one.

As the laser wavelength (532 nm) used in the experiment is farther out from the location of NLCs peak of absorption (~320 nm), the observed optical nonlinearities are related to present of Fe_3O_4 nps in NLCs.

Conclusions

In summary, the synthesized Fe_3O_4 nps were characterized by XRD and SEM, then these nps doped to NLC E7 to form FN. The third order nonlinear optical properties of FN were investigated at 532 nm using the low power single beam z-scan technique. The homeotropic alignment possess $\chi^{(3)}$ larger than that for homogeneous alignment.

Table

alignment	n_2(cm^2/W)	$\mathrm{Re}(\chi^{(3)})$	β(cm/W)	$\mathrm{Im}(\chi^{(3)})$	$\lvert\chi^{(3)}\rvert$
homogeneous	-5.2×10^{-7}	-119.8×10^{-6}	-6.5×10^{-2}	-20.2×10^{-6}	12.1×10^{-5}
homeotropic	0.55×10^{-5}	40.32×10^{-5}	1.5×10^{-1}	4.66×10^{-5}	40.5810^{-5}

Table1: Comparison of third order nonlinear optical parameters of a FN.

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