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Determination of Nonlinear Refractive Index of Zinc Phthalocyanine by Pump Induced Fizeau Interferometry

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ABSTRACT

Experimental measurement of intense light induced refractive index is demonstrated using Fizeau interferometer. Refractive index is induced in a specific region of the sample with intense light pulse of a linearly polarised, Q-switched Nd:YAG laser at 1064 nm wavelength. The respective change in optical path difference (OPD) is measured using Fizeau interferometer. The OPD so obtained is mapped in terms of change in refractive index in the area of interaction of pump beam and sample. The measured induced refractive index is then used to calculate thermo-optic coefficient of the ZnPc embedded polymeric sample.

Keywords: Fizeau interferometry; Thermo-optic coefficient; light induced refractive index

1. INTRODUCTION

Intense light induced refractive index is the fundamental process involved in several major nonlinear optical effects, including self-phase modulation, self-focusing, optical Kerr effect and optical phase conjugation. This effect is caused by third order nonlinear polarisation processes, and induced refractive index magnitude is proportional to the intensity of the incident light beam. From the application point of view, the effect of light induced refractive index can be utilised in development of many novel optical devices and techniques, such as data storage and processing, all-optical switching and steering and real time optical holography, etc. A no. of nonlinear optical characterisation techniques are known today and researchers are using these techniques for characterisation of new nonlinear materials. Among the various techniques, z-scan technique[1,2], optical Kerr gate[3,4], ellipse rotation[5], self-phase modulation[6] and nonlinear image processing[7] are generally used in measuring nonlinear refractive index of the nonlinear optical materials. Ashkin[8], *et al.* have observed the optically induced refractive index inhomogeneity in $LiNbO_3$, $LiTaO_3$ crystals using 632.8 nm and 514.7 nm wavelength lasers. Olbright[9], *et al.* have measured nonlinear refractive index of nonlinear materials using nitrogen laser pumped tunable dye laser in a Twymann Green interferometer configuration with a 5 ns pulse that has a resolution of 1/100 of a fringe shift. They measured the sample and reference fringe pattern intensity simultaneously using a 1024 channel linear diode array detector in a single shot measurement. Chen[10], *et al.* have measured the transient nonlinear refractive index in gases by developing a spectral interferometer capable of recording single shot records in Michelson interferometer configuration. Saifollah[11], *et al.* have used pump-probe technique for measuring nonlinear refractive index based on double grating interferometer. The pump beam has been aligned collinearly with the expanded parallel probe beam to distort the wavefront in the area of interaction. The method was finally applied for measuring the nonlinear refractive index of colloidal gold nano-particles and the nature of induced refractive index is found to be thermal. Tasnim[12], *et al.* have measured the nonlinear refractive index of 2,5 -dimethyleaniline (DMA) using Mach-Zehnder interferometer. The ratio of the measured input powers to the corresponding output powers is used to estimate the phase shift of nonlinear material.

Nonlinear refractive index of an optical material is a very important optical coefficient and it is require to be measured with high accuracy. The ability of an interferometer to measure nanometer level deviation makes it very sensitive and it can detect and measure very small changes in refractive index. Here, the measurement of induced refractive index is demonstrated based on the change in optical path difference (OPD) when the nonlinear optical material interacts with an intense light pulse. A Fizeau interferometer is used that has a resolution better than 1/600 of a fringe shift. Interference is a very sensitive and reliable phenomenon in measuring minute changes in OPD and is widely used in optical metrology for the surface accuracy measurements of optical components. The basic idea of testing inhomogeneity of optical materials like fused silica, BK7 glass etc, is used for the measurement of light induced refractive

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index change. Interference technique can detect refractive index variation of the order of 10^{-7} or less. Keeping in mind the inhomogeneity of the test sample, we can change the refractive index of the material by outside means in a specific region of the sample. The fringe distortion due to induced refractive index in the area of illumination can be measured and analysis will give the corresponding index variation. We have used this idea for measuring the nonlinear refractive index of PMMA based zinc phthalocyanine sample. This technique is quite reliable, sensitive and also the measurement is not affected by the pump laser beam profile.

2. THEORETICAL BACKGROUND

The intensity recorded on CCD image camera when the beams reflected from reference surface 1 and reference surface 2 of interferometer with intensity I_1 and I_2, respectively, interfere is given by[13]

$$I = I_1 + I_2 + 2\sqrt{I_1 I_2}\,Cos(\Delta\varphi) \tag{1}$$

where $\Delta\phi$ is the phase difference between the two interfering plane waves as shown in Fig. 1.

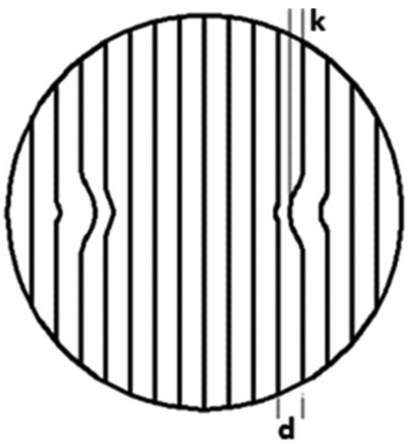


Figure 1. Interferogram with distorted wavefront.

If the maximum fringe deviation in the interferogram of the distorted wavefront is *k*, and the distance between two consecutive fringes is *d*, the optical path difference (OPD) is[14],

$$OPD = \left(k/d\right)\lambda \tag{2}$$

with λ as the wavelength of interferometer beam. Now the OPD due to induced inhomogeinity or induced refractive index change, Δn of the sample is given by

$$OPD = 2\Delta n t \tag{3}$$

where *t* is the thickness of the sample. The factor of 2 is introduced due to double pass of the wavefront through the nonlinear sample.

Comparing Eqns. (1) and (2), the induced refractive index in the sample is given by

$$\Delta n = \left(\frac{k}{d}\right)\left(\frac{\lambda}{2t}\right) \tag{4}$$

The standard relation between the induced refractive index Δn, and nonlinear refractive index n_2 which is of thermal origin, is given by[15]

$$\Delta n = n_{2(thermal)}\,|E_o|^2 \tag{5}$$

And also by[16],

$$\Delta n = \frac{dn}{dT}\frac{\Delta Q}{C_p \rho \upsilon} \tag{6}$$

here E_0 is the amplitude of the interacting optical energy of intense laser pulse, *dn* is the small index change induced by a small temperature variation of *dT* (*dn*/*dT* = thermo-optic coefficient), C_p is the specific heat, ρ is the density of the material, and υ is the volume of material heated by the light. ΔQ is the energy absorbed by the material per pulse, given by

$$\Delta Q = \alpha L I_o a \tau \tag{7}$$

here α is the absorption coefficient of the material and *a*, the cross-sectional area of the interacting pulse with τ as the duration of the laser pulse.

3. EXPERIMENTAL FINDINGS AND DISCUSSION

ZnPc dye is embedded in PMMA matrix by sol–gel method. Visually transparent ZnPc/PMMA (molar concentration 0.04 mM) sample is an isotropic media which is developed in the form of discs of thickness 1.62 mm and diameter 10 mm. These have been polished fine with cerium oxide on polyurethane pad after maintaining the parallelism of two surfaces less than 1 min of an arc. The second reference flat is used having surface accuracy better than λ/10 peak to valley error over 90 per cent of the surface. Flatness of the surfaces of sample is maintained with a peak error of less than 3λ (632.8 nm). Laser damage threshold of the material under investigation has been estimated to be 2.5×10^9 W/cm^2.

Figure 2 shows the experimental setup used. To excite the sample, a linearly polarised, Q-switched Nd:YAG laser (10 ns, 20 Hz) was used to generate pulses at 1064 nm wavelength. The peak power of the laser was 5 MW, which was focused by a converging lens of focal length 100 mm. The sample was placed at such a position that the Nd:YAG beam makes a small spot of 3 mm diameter on the sample at an angle of 25^o with the axis of interferometer. The interferogram has been recorded

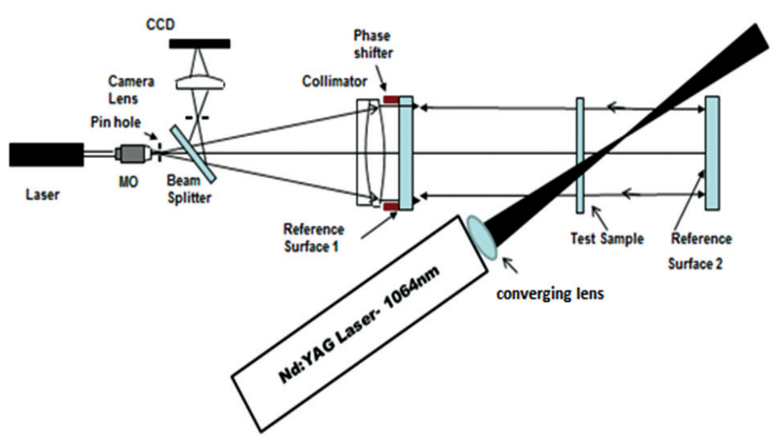


Figure 2. Experimental Setup for induced nonlinear refractive index measurement.

after placing the sample in between the two reference flats such that the wavefront is distorted while crossing the sample twice. Due to the high optical power density, nonlinearity arises in the area of interaction changing the refractive index of the material. In the area of interaction of pump laser with nonlinear sample, the fringe pattern changes because of the change in the OPD and consequently the fringes are distorted. The distortion in the fringes was measured and used to calculate induced refractive index of the material (Eqns. (2)-(3)). The two measured wavefronts (before and during Nd:YAG laser pulse firing) are subtracted from each other to give the respective change in the surface profile. The highly intense Nd:YAG beam induces an additional refractive index in the nonlinear sample under investigation. This induced refractive index changes the local profile of the wavefront in the area of interaction. The wavefront profile before and during the intense pulse triggering was recorded by CCD camera which are shown in Figs. 3(a) and 3(b), respectively. A minute change of 5 nm RMS over the full surface was observed because of the local variation in the small area of interaction. However, the robust PV (peak to valley) change was quite high because of the kinks in the profile. The average change in terms of PV was measured to be 25 nm, which was quite observable and sufficient to extract the information about the nonlinear material. The effect of induced refractive index is shown in Figs. 3(c) and 4 in 2-dimensional and 3-dimensional views. The interferometer was operated in continuous mode to detect every significant change in the double pass wavefront.

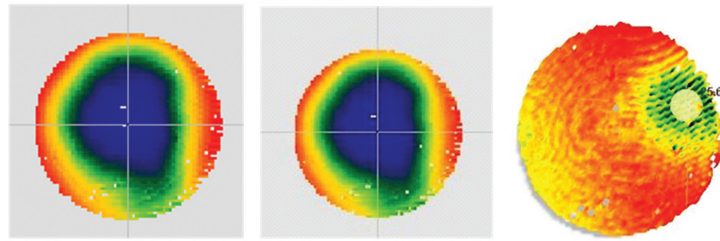
The induced refractive index was deduced from Eqn. (4) by measuring the fringe distortion in terms of peak to valley difference in the profile. The induced refractive index was then used to calculate nonlinear refractive index n_2 and thermo-optic coefficient using Eqns. (5) and (6), respectively. The results are summarised in Table 1.

4. CONCLUSION

A new method has been demonstrated for measuring induced refractive index in the nonlinear media using Fizeau interferometer. The nonlinear refractive index of the material is deduced using standard relations. The main contribution in the nonlinear refractive index is of thermal origin. The thermo-optic coefficient of ZnPc sample is also calculated. The technique is quite sensitive and very accurate because of the interferometer associated with it.

Table 1. Experimentally deduced nonlinear optical coefficient of ZnPc

Wavelength (nm)	Freq. (Hz)	Peak Intensity (I_0) W/cm^2	Induced Ref. Index	Nonlinear Ref. Index (cm^2/W)	Thermo-optic Coefficient (dn/dT) K^{-1}
1064	20	1.02×10^8	1.58×10^{-5}	15.5×10^{-14}	0.5891×10^{-5}



(a) (b) (c)

Figure 3. (a) profile of the wavefront before triggering with intense laser pulse, (b) during excitation with the intense laser pulse, (c) 2-dimensional contour profile after subtracting (a) and (b).

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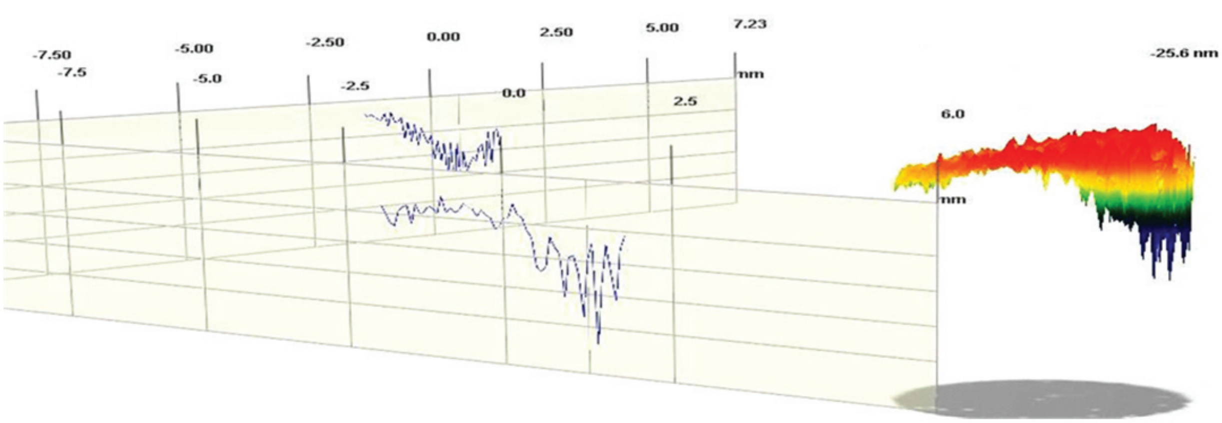

Figure 4. 3D profile of the subtracted wavefronts.

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In the current study, he prepares the framework for the study, result analysis and manuscript writing.

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He has been associated in reviewing the entire study and has edited the manuscript.