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Effect of viscosity on the far-field diffraction pattern of spatial self-phase modulation

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Effect of viscosity of a liquid on the far-field diffraction pattern due to spatial self-phase modulation is investigated. Chlorophyll extract in acetone at different concentrations is used as the low viscous nonlinear samples and commercial engine oils, synthetic and natural. It is also used as high viscous media. In the case of chlorophyll solution, the self-induced diffraction ring pattern in the far-field is a series of circular rings at the beginning; but later it is transformed to half circular symmetry. But for the high viscous engine oils we have observed circular fringes and there is no time dependent structural variation in the pattern. The coefficient of the intensity dependent nonlinear refractive index for all the liquid samples is evaluated.

Keywords: self phase modulation, nonlinear refractive index, chlorophyll, engine oil.

1. Introduction

Light intensity at the centre of a Gaussian laser beam is much larger than that at the edge. As a result, when this beam is used to heat a liquid sample, the change in refractive index of the medium is maximum at the centre of the illuminated spot and decreases radially as a function of distance from the centre of the beam. This light-induced spatially inhomogeneous refractive index profile can have considerable effects on laser beam propagation in a nonlinear medium, which leads to a well-known self-action effect phenomena known as spatial self-phase modulation (SSPM) as observed in several systems, such as, liquid crystals, polymers, nanoparticles, crystals, tea, oils, *etc*. [1–10]. SSPM can be of electronic or thermal origin and can be observed with a high power pulsed or low power continuous wave laser [11–18]. In the absence of any convective fluidic flow within the heated volume, the self-induced diffraction pattern may be a set of concentric circular rings. But local heating of the liquid can initiate convective [19–23] flow and these rings can be distorted as explained theoretically and experimentally by Karimzadeh [24, 25]. In his paper, the behavior of the diffraction ring patterns is interpreted theoretically based on the Fresnel–Kirchhoff integral and the analytical solution of the heat transfer equation. According to his model, laser heating is caused by coupled conductive and convective transport of thermal energy. At the beginning, circularly symmetric diffraction ring patterns are observed in the far-field, but as time elapses, the convective effects become dominant and circularly asymmetric diffraction ring patterns are formed.

A high viscous and a low viscous nonlinear media are used in this study to demonstrate the absence and presence of a self-induced natural convection flow on the SSPM diffraction pattern. Chlorophyll at different concentrations dissolved in acetone is taken as the low viscous nonlinear media. Chlorophyll is the green pigment found in plant leaves and is known to possess high nonlinear optical property [2]. Petroleum-based and synthetic engine oils are used as high viscous media, whose nonlinear optical properties are not known. We have chosen these oils because preliminary investigations showed that these oils exhibit SSPM when irradiated with the laser beam.

2. Theory

Under the influence of intense laser beam, the intensity dependent refractive index is given by

$$n = n_1 + n_2 I \tag{1}$$

where n_1 is the linear refractive index, I is the intensity of the light, and n_2 is the coefficient of the nonlinear refractive index. In this study we have evaluated the value of n_2 by counting the spatial self-phase modulation rings.

The theory of spatial self-phase modulation is discussed in detail elsewhere and hence we give only a brief description here [26, 27]. Self-phase modulation is a consequence of the difference in the nonlinear refractive index at different radial positions of a beam due to its non-uniform spatial intensity profile. The variation of the refractive index induces changes in the optical path of each part of the beam. SSPM phenomenon caused by a Gaussian beam can be seen as diffraction of light through a circular aperture, which introduces a phase shift depending on the local light intensity. The circular aperture corresponds to the beam width on the sample, while the phase shift profile is due to the nonlinear response of the medium. For an incident Gaussian beam whose electric field is written in the form

$$E_\mathrm{r} = E_0 \exp\left(-\frac{r^2}{w^2}\right) \exp\left(-\frac{ikr^2}{2R}\right) \tag{2}$$

the transmitted intensity given by the Fraunhofer diffraction integral can be written as

$$I(\theta) = \left(\frac{2\pi}{D}\right)^2 I_0 \left| \int_0^\infty r\,\mathrm{d}r J_0(k\theta r) \exp\left(-\frac{r^2}{w^2}\right) \exp\left\{-ik\left[\frac{r^2}{2R} + B\exp\left(-\frac{2r^2}{w^2}\right)\right]\right\} \right|^2 \tag{3}$$

where E_r is the electric field at the radial distance r from the center of the beam, E_0 denotes the electric field at the focus, w is the beam width, R is the radius of curvature of the wave front, and $k = 2\pi/\lambda$, r is the radial distance, D is the distance from the sample, θ is the diffraction angle, J_0 is the zeroth order Bessel function, $B = \Delta n L$, where Δn is the maximum induced variation of the refractive index and L is the thickness of the sample.

The nonlinear phase shift induced by a plane wave is given by

$$\varphi_{\mathrm{nl}}(r) = \frac{2\pi}{\lambda}\int_0^L \delta n(r, z)\mathrm{d}z \tag{4}$$

Here δn is the refractive index variation experienced by the incident wave. The nonlinear phase shift contribution is related to the intensity-dependent refractive index of the material. If we assume the Kerr nonlinearity, the refractive index changes can be represented as

$$\delta n(r, z) = n_2 I(r, z) \tag{5}$$

The magnitude of the nonlinearity can be found out from the number of the principal SSPM rings. Hence we can see that the nonlinear phase shift is a function of the light intensity, which in turn is Gaussian in shape, φ_{nl} can have same value for two different values of r. It means that these portions of the wave front travel in the same direction and can interfere. The interference pattern will appear as bright and dark rings in the far field. The maximum phase shift $\delta\varphi_0$ can easily be related to the number of rings N by,

$$\delta\varphi_0 = 2\pi N \tag{6}$$

From the expression for a nonlinear phase shift, we can get the maximum phase shift in a sample induced by a Gaussian planar wave as

$$\delta\varphi_0 = \frac{2\pi}{\lambda}\delta n_0 L \tag{7}$$

Equating the two equations for $\delta\varphi_0$ and substituting for δn_0, we get

$$N = \frac{n_2 L}{\lambda} I_0 \tag{8}$$

From the slope of the number of rings in the diffraction pattern *vs*. intensity plot, we can easily determine the coefficient of the nonlinear refractive index. The coupled conductive-convective transport theory by KARIMZADEH [24, 25] only accounts for the asymmetric nature of the diffraction ring pattern and hence the previously discussed non-convective theory can be used to estimate the coefficient of the nonlinear refractive index of such samples which show convective heat flow.

3. Experiment

Dried and powdered Indian tea-plant leaf from Western Ghats is boiled in water for several minutes to dissolve water soluble chemicals. The water is then strained away and the residue is collected. The same procedure is repeated several times until the residue is free from all water soluble components. The tea powder is placed in an oven and dried at 60°C until its moisture content is completely removed. A small amount of this tea powder is taken in a glass flask and sufficient amount of acetone is added to it. Now the chlorophyll in the tea leaf is extracted in acetone and a green colour solution is obtained. Different concentrations of chlorophyll extract are prepared by diluting the stock solution with acetone and each concentration is characterized by its optical absorbance at 532 nm.

Two different engine oils, namely, petroleum-based regular engine oil and synthetic engine oil are used as high viscous media for the SSPM studies. A continuous wave diode pumped solid state laser emitting 50 mW at 532 nm is used as the light source. The intensity of the laser beam is controlled using a neutral density filter set. A power meter is used to measure the laser power. The laser beam is focused using a plano-convex lens of focal length 10 cm to a spot size of 47.5 μm. Chlorophyll samples are taken in a 1.5 mm cuvette and the engine oil samples are taken in a 3 mm cuvette. Diffraction ring pattern due to SSPM is observed on a screen placed about 2 m from the sample. Temporal development of the ring pattern is recorded using a digital camera operating at 30 frames per second in the video mode.

4. Results and discussion

The laser beam is blocked using an electromechanical shutter and suddenly the shutter is opened. Video of the far-field diffraction pattern is recorded. Free software VirtualDub is used to generate frames from the video. The number of rings is counted directly from the screen as well as the free software Tracker is used to count the number of rings from the still pictures extracted from the video. The procedure is repeated for different laser intensities by changing the combination of neutral density filters. The entire experiment is repeated for different samples, namely, the petroleum-based and synthetic engine oils and for three different concentrations of chlorophyll in acetone.

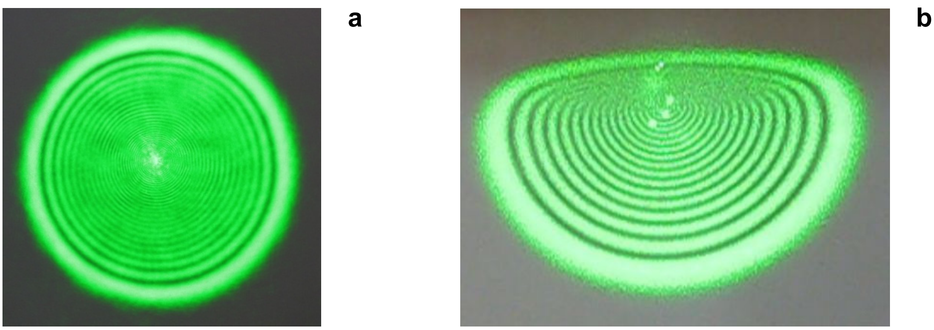

Fig. 1. The fully grown far-field circular diffraction pattern from the petroleum-based engine oil (**a**). The asymmetric diffraction pattern from the chlorophyll in acetone (**b**).

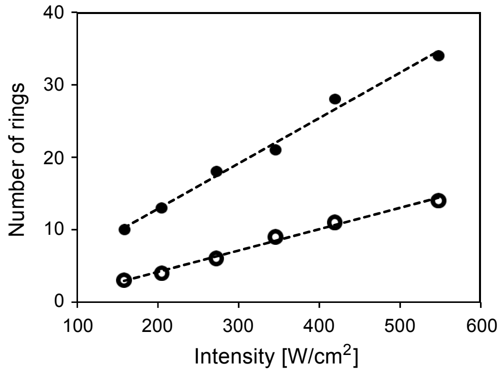

Fig. 2. Intensity *vs*. number of diffraction rings plot for petroleum-based (filled circles) and synthetic (open circles) engine oils.

T a b l e. Coefficient of nonlinear refractive index of the chlorophyll and engine oil samples.

Sample	Absorbance at 532 nm	Threshold intensity [W/cm^2]	Coefficient of nonlinear refractive index [cm^2/W]
Chlorophyll	0.13	28	5.37×10^{-6}
Chlorophyll	0.29	16	9.38×10^{-6}
Chlorophyll	0.5	10	13.6×10^{-6}
Engine oil (petroleum-based)	–	32	11.1×10^{-6}
Engine oil (synthetic)	–	70	5.22×10^{-6}

The observed circular symmetry in the far-field diffraction patterns of engine oils is shown in Fig. 1**a**. It is observed that the number of diffraction rings increases linearly with the light intensity as shown in Fig. 2. The coefficient of the nonlinear refractive index is calculated using the equation given in the theory section and is tabulated in the Table. Petroleum-based engine oil is found to be more nonlinear than the synthetic oil. Chlorophyll in acetone at three different concentrations is used for the studies. From the video frames it is observed that at first the self-induced diffraction ring pattern in the far-field is a series of circular rings; but as time elapses, quickly the rings changed to half circular symmetry as shown in Fig. 1**b** (only the final frame is shown). This is attributed to the convection effect in the liquid sample. The number of rings *vs*. the light intensity plot is shown in Fig. 3. At very low intensities we have observed a deviation from the linear relationship and those values are not used in the graph. The coefficient of the nonlinear refractive index is calculated and tabulated in the Table. It can be seen that the coefficient of the nonlinear refractive index increases with the concentration of the chlorophyll.

The measured n_2 values of chlorophyll are of the same order as those of plant extracts and organic dyes in solvents [28, 29] and the measured n_2 values of engine oils are one order greater than those of pure castor oil and one order less than those of castor oil and palm oil with nanoparticles dispersed in it [3, 10]. The chlorophyll concentrations

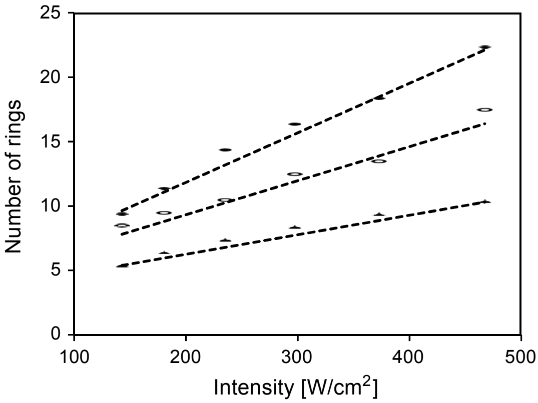

Fig. 3. Intensity *vs*. number of diffraction rings plot for different concentrations of chlorophyll in acetone (filled circles – absorbance equal to 0.50, open circles – absorbance equal to 0.29, and filled triangle – absorbance equal to 0.13).

are selected such that the nonlinear refractive index is close to that of the engine oils to make sure that this asymmetric shape of the ring pattern is not due to a completely different (too high or too low) nonlinear refractive index compared to that of the engine oil. Density of acetone is 791 kg m^{-3} and that of engine oil is less than 900 kg m^{-3}. Viscosity of acetone is 0.306×10^{-3} N s m^{-2} and that of the engine oil under investigation is in the range of 70×10^{-3} to 90×10^{-3} N s m^{-2}. Since convective heat flow is inversely proportional to the viscosity, low viscosity of acetone causes increased convective flow in the acetone sample compared to the engine oil sample, which is in agreement with the theory of convective heat flow within the local heated volume. According to this theory, the asymmetric ring pattern due to SSPM could be an indication of the onset of convection caused by the laser-induced heating [24, 25]. Absorption of light energy and subsequent non-radiative deexcitation results in temperature gradient and hence density gradients in the liquid sample. Considering the Gaussian beam profile, temperature will be highest at the axis of the illuminating cylindrical volume and decreases radially. Considering the upper half of the illuminated horizontal cylindrical volume, the density decreases with an increase in distance from the base of this upper half cylinder, very similar to that of a liquid heated from bottom. The natural convection in the liquid causes the heated liquid to move upwards and its original place is occupied by cool liquid from the immediate surroundings. Considering the lower half of the illuminated horizontal cylindrical volume, maximum temperature and hence minimum density of the liquid is at the top of this horizontal half cylinder and hence no significant natural convective motion occurs in this bottom half. Consequently, the far-field diffraction ring pattern of the Gaussian laser beam propagating through the liquid shows an asymmetric shape in the vertical direction, and looks like squeezed from the top instead of circular fringes.

The ring pattern from the engine oils is found to show the circular symmetry throughout its development and until it is fully grown and hence no further analysis is required.

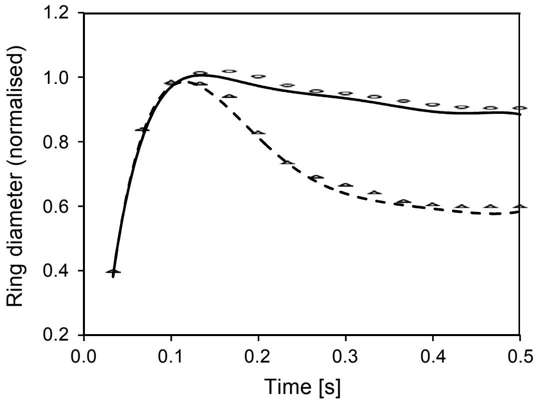

Fig. 4. Ring diameter *vs*. time plot for chlorophyll in acetone (absorbance equal to 0.5). Open circles for horizontal diameter and open triangle for vertical diameter. Ring diameter is normalized with the maximum horizontal diameter.

But in the case of a chlorophyll sample, first few frames of the video show perfectly circular fringes but later it became asymmetric. The diameter of the ring pattern along the horizontal and vertical direction is measured and the graph connecting the time *vs*. the diameter is shown in Fig. 4. The diameter value for chlorophyll in acetone having an absorbance of 0.5 is shown in a normalized scale using the maximum diameter, which is along the horizontal direction. A slight decrease in the horizontal diameter is also accounted for by the convective flow in the upper half of the illuminated volume. Similar pattern is also observed for the other two chlorophyll concentrations.

5. Conclusion

The effect of viscosity on the nonlinear optical behavior of two different types of samples having similar values of nonlinear refractive index is studied. Results are in agreement with the existing theories of convective heat flow in the heated volume for the low viscous liquid, whereas no such effect is visible in highly viscous engine oils. From the analysis of the ring pattern originated due to spatial self-phase modulation, the coefficient of the nonlinear refractive index of the samples is also estimated. Please note that this method does not give the sign of the n_2 value, but for most of the liquids, it is negative. Petroleum-based engine oil is found to possess very high optical nonlinearity, almost double than that of synthetic engine oil, and both engine oils are free from convective heat flow within the laser beam heated volume. Hence, considering the low cost, high viscosity and high optical nonlinearity, petroleum-based engine oil is a good candidate for nonlinear optical applications in the low power regime.

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