Observation of the far field diffraction patterns of divergent and convergent probe laser beam in DAZA polymer

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We report the observation of the far field self-diffraction rings patterns in Poly(azaneylylidene-acylene) "DAZA" polymer under the influence of the dual laser beams. The incident pump laser beam was varied as: 10mW, 20mW, 30mW, 40mW and 50mW in order to study the effect of the laser power on the number of the rings. Also, the difference between the ring shapes in both sides with respect the beam curvature was observed. The Fresnel-Kirchhoff diffraction formula was applied to perform simulation of the self-diffraction rings patterns with different laser power used as a pump power at two sample positions before and after beam focal point with respect to the geometrical pumping. Our results are in good agreements between the experimental data and the propose model based on the Fresnel-Kirchhoff diffraction formula.

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1. Introduction

Spatial self-phase modulation (SSPM) is considered a nonlinear phenomenon, that is observed in far field diffraction ring patterns, when Gaussian laser beam is focused into absorbing thermal nonlinear (NL) medium [1-3]. So, SSPM effect can be utilized as very important tool to characterize new NL materials for possible use in future optoelectronic devices [4-8]. The observed patterns have a series of concentric rings. The number of the rings depends on the sample position with regarding to the focal point (z=0), as well as the incident laser energy [9]. It was reported that the central area (dark or bright) of the rings is changed according to the sample position, whether it in front or behind the focal point position [10]. These important features can be clarify as result of the interaction of strong laser light with NL medium, then the induced distribution heating is generated inside the NL medium due to the conduction and convection heat currents [11]. When a low power laser beam was used as excitation source, there would be no diffraction rings patterns in the experiment. So, we need two laser sources (pump and probe beams) to overlapped together, this leads to have a cross-phase modulation (XPM) technique, which resulting in generating diffraction rings with low laser intensity. With the increase of laser beam power, the nonlinear phase shift will be changed by the spatial cross-phase modulation (SXPM)[12]. In both cases, SSPM and SXPM, the convection heating effect was minimized by introducing the focused laser beam in parallel way with the acceleration gravitation forces into the sample cell, therefore the symmetric diffraction patterns are formed at far field, and vice versa when the incident laser beam moved toward the sample cell in vertical direction with the gravity forces, the distortion rings patterns will be observed [9]. However, the concepts of the SSPM effect based on the experimental and theoretical studies have already been reported in full details elsewhere [13-16].

Recently, SSPM effect has been reported in different materials, such as: liquid suspensions of graphene [17, 18], black phosphorus [1], carbon nanotubes [19], dispersive suspension of graphite flakes[20], and some others organometallic molecules[16, 21, 22]. Also, the NLO properties of some inorganic pigment, Azophloxine and Alizarin Red S were studied[23-25].The diffraction rings patterns in hot atomic sample were studied, the results indicated that the formation of diffraction rings patterns will be changed at various sample positions with respect to Z=0 [26]. It has been reported of using the SXPM (a dual-color) technique, in order to study all optical switching effect in the MoS_2 compound[27], and in the $CsPbI_3$ QDs, $CsPbBrxI_3$ [28].

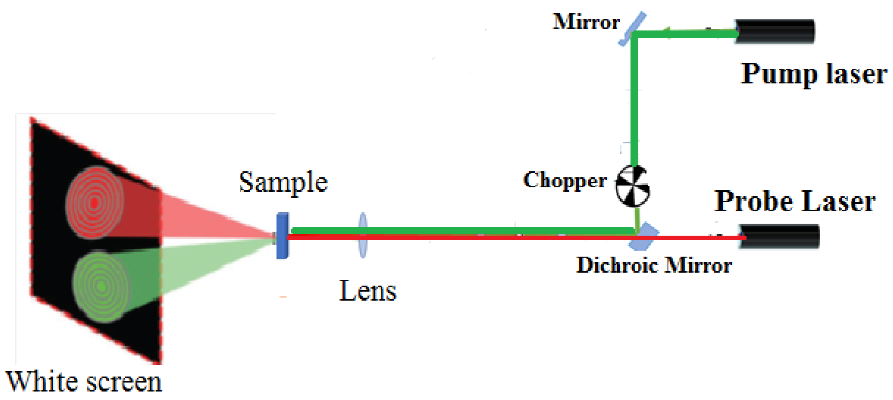
Polymers are considered very important class of organic materials, as well as they exhibit fast nonlinear optical response times. Polymers materials have been reported in large number of studies, for observing the far field diffraction rings patterns in order to fabricate optical devices [29-32].

The Fresnel–Kirchhoff diffraction theory was utilized to investigate the influence of the front wave curvature [R(z)] on the far-field rings patterns [4, 33]. The theoretical data has shown the formation of diffraction rings patterns due to the interaction of the Gaussian laser beam with NL media (self-focusing or self-defocusing media) [28, 34, 35]. The nonlinear refractive index (n_2) and the 3rd nonlinear susceptibility of the molecules can be determined by the relationships between the number of SSPM rings and the laser intensities[2].

In this work, the nonlinear optical properties of the new DAZA polymer were investigated via SXPM technique using green laser as a pump beam (CW laser, λ=532 nm) and diode red laser as probe beam (λ= 632.8 nm). The diffraction ring patterns at far field were observed using the SXPM technique, then they were numerically simulated using the Fresnel-Kirchhoff formulism in order to confirm the experimental data.

2. Experimental techniques

The DAZA polymer was already prepared and fully characterized using different spectroscopic techniques[36]. Schematic 1 shows the spatial cross-phase modulation (SXPM) experimental setup. Two laser beams were directed toward into the sample cell with a 10 cm convex lens. The Gaussian TEM_{000} diode laser system (λ=532nm) was used as a pump laser with power up to 100 mW, and the second laser used as probe beam (He-Ne laser λ= 632.8nm) with power up to 1mW. One mirror was used in front of the green pump beam and Dichroic was fixed before the sample to make mode-match configuration between the pump and probe beams. Beyond the sample cell, the probe and pump beam signals are directed into white screen (about 1 m) in order to capture the diffraction rings patterns with help of digital camera for further processing. Optical chopper was used in front of the pump laser beam to make optical modulation; and the sample was fixed on translational stage. The DAZA polymer has wide absorption range from 280 to 700 nm[37].



Schematic 1. The spatial cross-phase modulation (SXPM) experimental setup (color online)

3. Results and discussion

To generate the self-diffraction rings at far field by SSPM modulation; a strong laser beam should be used. Usually, the self-diffraction rings will not be observed in the case of using low power laser (probe laser). But, the spatial cross-phase modulation (SXPM) technique can be used to observe self-diffraction rings, when a strong laser beam is used simultaneously with low power laser beam. This process is performed as results of overlapping between the strong and low power laser beams.

We were able to use the DAZA polymer as NL medium (sample) and utilizing the SXPM technique in order to investigate the far field self-diffraction rings at different laser powers for two sample cell positions. Fig. 2 shows the images of self-diffraction rings patterns of the probe laser beam using the spatial cross-phase modulation (SXPM). The incident pump laser beam was varied as: 10mW, 20mW, 30mW, 40mW and 50mW in order to see the influence of the laser power on the number of the rings. At each laser power value, the sample cell was fixed in front the focal point position (z =-1cm) and after the focal point position (z = +1 cm) in order to investigate the influence of the beam curvature on the observed diffraction rings patterns, which show the difference between the ring shapes in both sides. As, it was mentioned earlier, the aim of this measurements to confirm simple fact; a weak laser beam can be modulated in consequence with strong pump laser beam. Our results have shown the brightness, diameter and number of diffraction rings increase with increasing the pump laser power, due to creating of thermal lensing and hence presence of larger phase shifts. With respect to the influence of the beam curvature (R), and our sample is self-defocusing medium, this means the non-linear phase shift is $\Delta\phi<0$, there will be two cases: the first case is $\Delta\phi$ <0 and R< 0 at Z= -1 cm, the observed rings become thicker as the power is increased. The width of the rings is larger for the outer rings than it is for the inner ones. The second case is $\Delta\phi$<0 and R> 0, the sample was positioned at z =+1 cm, one can observe that the innermost part of the rings is diffuse and that a brighter external ring is generated for this case. The number of generated rings varies linearly with the laser powers.

The mean feature of the diffraction rings is asymmetrical, it is seem that the upper half (vertical direction) of the ring is collapsed with respect to the lower part (horizontal direction). This is called "the distortion effect" which is due to the gravity and convection currents. They are in opposite directions on the vertically positioned of the sample cell in relative to pump beam direction, which produced a series of concentric circles, each with flattened tops, the distortion effect was explained in full details elsewhere [9].

As, the experimental results show asymmetric diffraction ring patterns due to the presence of convective effects. We have obtained a numerical results for the far-field intensity distribution of a Gaussian laser beam propagating through the DAZA polymer using the integral formula of Fresnel-Kirchhoff diffraction [33]:

$$I(x',y',t)=\left| \begin{array}{l} U_0 \frac{i\pi\omega^2}{\lambda d}\exp\left(-ikd\right)\exp\left(-\frac{\alpha L}{2}\right) \\ \times\int_{-\infty}^{+\infty}dx\int_{-\infty}^{+\infty}dy\ \exp\left(-\frac{x^2+y^2}{\omega^2}\right) \\ \exp\left[i\left(-k\frac{x^2+y^2}{2R}+\Delta\phi\left(x,y,t\right)\right]\right. \\ \times\exp\left(\frac{-ik\ (xx'+yy')}{d}\right) \end{array} \right|^2 \quad (1)$$

where x' and y' are the Cartesian spatial variables in the detector plane and d is the distance between the exit plane of the sample and the detector plane. The phase shift $\Delta\phi(x,y,t)$ is given by[33] :

$$\Delta\phi(x,y,t)=\frac{\theta}{2t_c}\int_0^t\frac{dt'}{1+\frac{2t'}{t_c}}\times\left(\begin{array}{l}\exp\left(\frac{-2[(x-v_xt')^2+y^2]}{[(1+\frac{2t'}{t_c})\omega^2]}\right)\\ -\exp\left(\frac{-2[(v_xt')^2}{[(1+\frac{2t'}{t_x})\omega^2]}\right)\end{array}\right) \quad (2)$$

The t_c, D, and θ terms are defined as follow: $t_c=\omega^2/4D$ is the characteristic heat diffusion time, $D=k/\rho c_p$ is the thermal diffusivity of the sample, and θ is the on-axis phase shift [$\theta=\frac{(dn/dT)\alpha P L_{eff}}{\lambda k}$], where (dn/dT) is the thermo-optic coefficient and P is the laser power.

The term of heat convection velocity (v_x) was calculated using the following relation 3[33] :

$$v_x=\frac{\beta\, g\,[\Delta T]_{\max}\,\pi\, h^2}{16\,\mu} \quad (3)$$

where g is the acceleration due to gravity, β is the thermal expansion of the medium, μ is the liquid viscosity, h is the minimum distance from the center of the beam to the meniscus, and the ΔT is the maximum change in temperature. As a result of interaction between Gaussian laser power and NL medium, with presence of the convection effect component, the distribution of temperature ΔT(x,y,t) inside the NL medium is given [33]:

$$\Delta T(x,y,t)]=\frac{\alpha\ p}{\pi\,\rho\,c_p}\left[\begin{array}{l}\int_0^1\frac{d\,t'}{8Dt'+\omega^2}\\ \times\exp\left(\frac{-2\,[(x-\upsilon_x t')^2+y^2}{[8Dt'+\omega^2]}\right)\end{array}\right] \quad (4)$$

(All the terms in the equations 1-4 were define in reference[33])

Using equation 1, the simulated asymmetric diffraction patterns in the presence of convention effect component of DAZA polymer were shown in Fig. 3. The mean features of the numerical diffraction rings are increased and the size of rings is larger with increase the pump power. Also, the center of the rings is look like a bright spot when R<0 and dark spot when R>0. Then, the diffraction patterns are formed and the number of the rings is increased with laser powers. However, the vertical asymmetry of the intensity profile about the central horizontal position has increased. The presence of distortion is a result of convective heat transfer component in the sample. The distortion phenomenon in the diffraction rings was already discussed in full details elsewhere [38, 39].

Finally, we can say that there is a good agreement between the experimental and numerical results as both Figs. 2 and 3, they show asymmetric diffraction ring patterns for a Gaussian laser beam at the far-field.

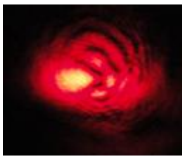
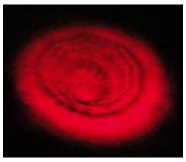
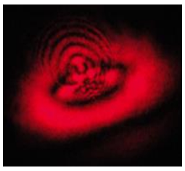
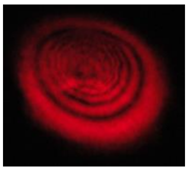
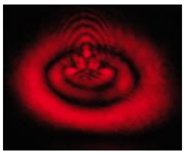
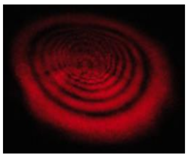
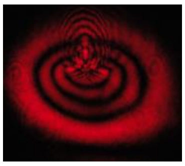
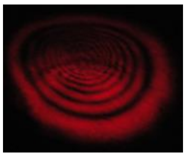
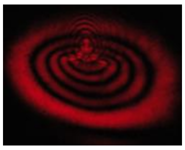
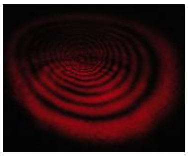
Sample position		Pump Laser Power (mW)
Z = - 1 cm	Z = +1 cm	
		10
		20
		30
		40
		50

Fig. 2. Experimental images of asymmetric diffraction rings at two sample positions (z=+1 cm and z=-1 cm) with different laser powers of probe laser wavelength, λ= 632.8 nm (color online)

4. Conclusion

We present the results of experimental and numerical study of self-diffraction rings patterns using the spatial cross-phase modulation (SXPM) technique of DAZA polymer. Our results have shown that the formation of the self-diffraction rings patterns depends on the positions of the sample with respect to the waist of the Gaussian laser beam and the input laser powers. Our studies on DAZA polymer have shown that the DAZA polymer is a self-defocusing material, this means that the phase shift $\Delta\phi<0$. When the positive curvature radius $R(z) > 0$, the central spot is dark and a central bright spot for negative curvature radius $R(z)<0$. The number of rings in the diffraction patterns increases with increasing the input pump power. Also, our results have shown good agreement between the experimental and the numerical study.

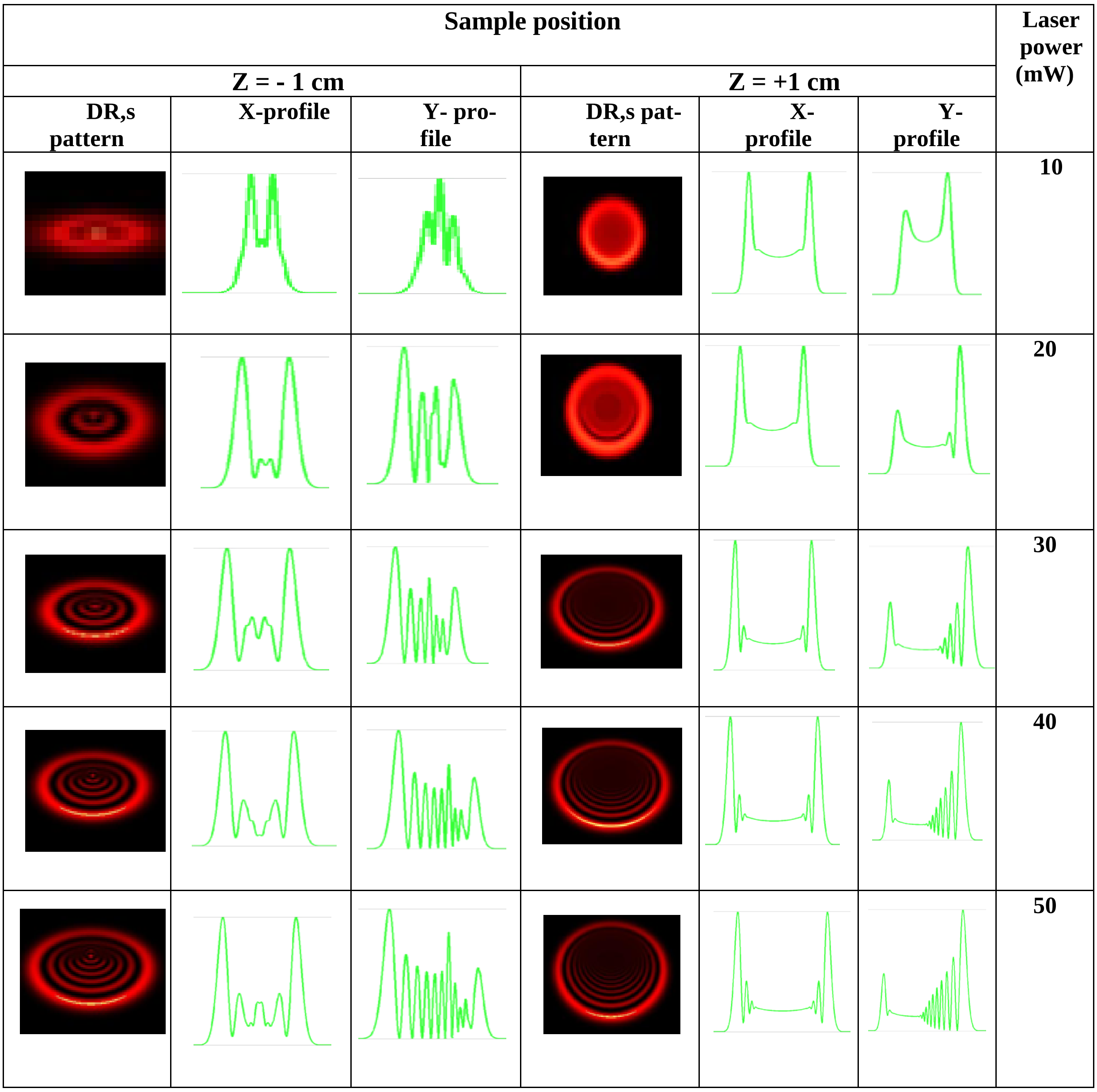


Fig. 3 Calculated of asymmetric diffraction rings at different laser powers at two sample positions (z=+1cm and z=-1cm) with probe laser wavelength, λ= 632.8 nm (color online).

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