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Laser-induced pattern formation from homogeneous polyisoprene solutions*

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This paper reports that the pattern formation in homogeneous solutions of polyisoprene in toluene saturated with C_{60} induced by a continuous-wave visible laser is observed experimentally. The transmitted beam patterns change with the increase of the laser irradiation time. In the initial phase, the patterns with concentric ring-shaped structure are formed. In the end, the patterns become speckle-shaped. The incubation time of the transmitted beam widening is inversely proportional to the laser power density and solution concentration. The pattern formation results from the optical-field-induced refractive index changes in the solutions, but the mechanism of optical-field-induced refractive index changes in the polymer solutions needs to be further studied.

Keywords: laser-induced, pattern formation, polyisoprene
PACC: 4265J, 4270J

1. Introduction

The interactions of light with matter have extreme scientific and technological importance. Radiation forces from a laser beam can be very strong because of the beam's coherent and highly directional properties. It is therefore possible to use a focused laser beams to accelerate, decelerate, steer, manipulate, cool, or trap small particles.[1−6] This has opened a fascinating area of laser physics. Optical-field-induced refractive index changes can have an appreciable effect on laser propagation in a nonlinear medium. A laser beam can induce self-focusing, self-trapping and self-phase modulation in Kerr media, and thermal blooming and photo-thermal effects in photo-thermal materials,[7−11] as well as cross-linked polymerization in some polymer materials.[12,13] Scattered and transmitted light can reflect the materials structure and dynamics. In 2002 R. Sigel *et al* reported pattern formation in homogeneous polymer solutions induced by a continuous-wave (CW) visible laser.[14] This optical effect lacks at present a fully satisfactory explanation, and it could potentially lead to photorefractive, microoptics, and nanotechnology applications.

In this paper, the pattern formation in homogeneous solutions of 1,4- polyisoprene (PI) in toluene saturated with C_{60} induced by a CW visible laser beam is observed in the experiment. The various pattern images are taken with the increase of the laser irradiation time and with the change of the focal point positions. The effects of the laser wavelength and power density, as well as solution concentration on the pattern formation are obtained. The experimental results are discussed and analysed.

2. Experimental setup

The experimental setup is shown in Fig.1. After passing an aperture with diameter of 1 mm, a CW visible laser beam with a Gaussian-like transverse profile is focused by a lens with a focal length of 8 cm and into the rectangular optical cell (1 cm×1 cm) containing the polymer solution. Samples are a homogeneous semidilute solution of 1,4-PI [–H_2C–$C(CH_3)$ = CH–CH_2–]$_N$ ($N \sim 11760$) in toluene and that of 1,4-PI in toluene saturated with C_{60}. The transmitted beam spot from the sample cell is observed on a screen and imaged by a charge-coupled device camera. The distance between the screen and the output window of

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the cell is about 3 m.

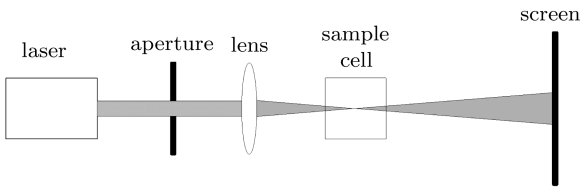


Fig.1. Experimental setup.

3. Experimental results

For a 0.0083 g/ml solution of 1,4-PI in toluene, when a laser beam with the wavelength of 632.8 nm is used and its power is less than 2.0 mW, the transmitted beam does not widen; when the laser power is more than 2.0 mW, the transmitted beam widens extremely slowly, and its pattern becomes concentric ring-shaped. Once the patterns are formed, the solutions have memory effect, and it vanishes extremely slowly for about several days. For a 0.0083 g/ml solution of 1,4-PI in toluene saturated with C_{60}, the laser power causing the transmitted beam to widen decreases obviously, and the beam pattern in the solution of PI in toluene saturated with C_{60} changes faster than that in the solution of PI in toluene without C_{60}. The solutions of PI in toluene saturated with C_{60} have memory effect like that of PI in toluene. The images of the transmitted beam patterns observed on the screen for various times are shown in Fig.2. The initial laser beam shown in Fig.2(a) has a Gaussian-like transverse profile. About 0.1s after the laser radiates the sample, the transmitted beam widening is observed. At 1 s, 20 s, 130 s and 368 s, the images of the patterns taken are shown in Figs.2(b)–(e) respectively. At 654s, the pattern shown in Fig.2(d) becomes speckle-shaped.

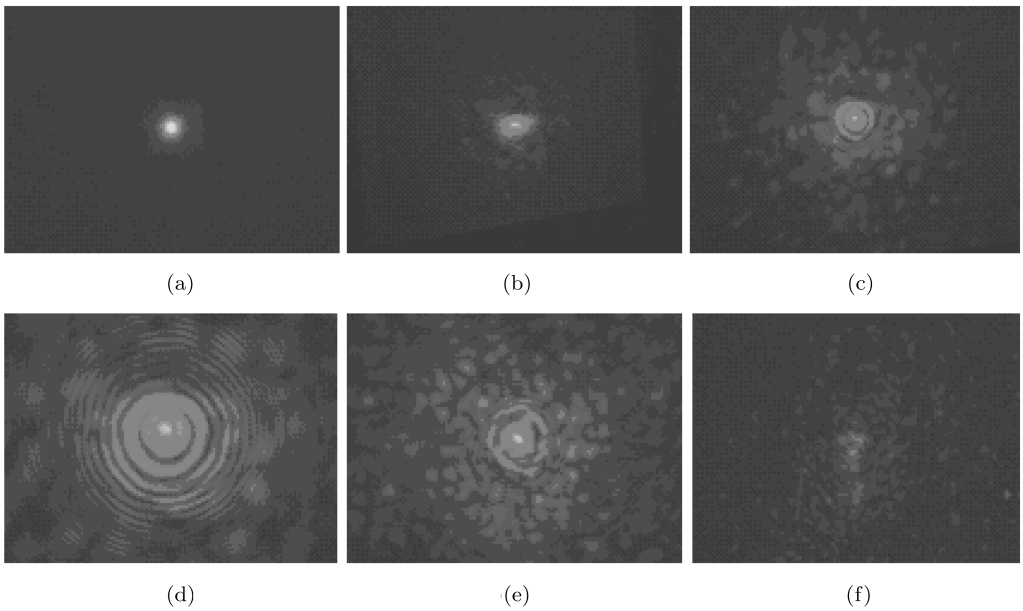
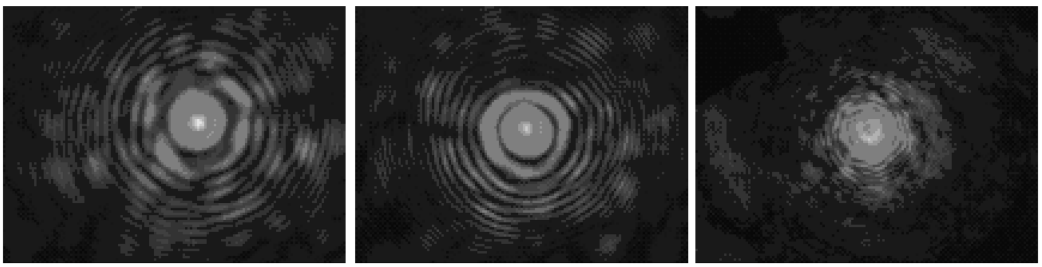


Fig.2. Images of the beam patterns observed on the screen for various times: (a) initial laser beam; (b) at 1 s; (c) at 21 s; (d) at 130 s; (e) at 368 s; (f) at 654 s.

For a 0.0170 g/ml solution of 1,4-PI in toluene saturated with C_{60}, the incubation time (from 0.1 s to 300 s) for the transmitted beam widening is found to be inversely proportional to the laser power (from 0.3 mW to 2.2 mW). For a solution of 1,4-PI in toluene saturated with C_{60} and with the use of a 2.5 mW laser beam, the incubation time (from 0.1 s to 110 s) is found to be inversely proportional to the concentration (from 0.0060 g/ml to 0.0230 g/ml) of PI in the solution. For a low concentration solution and with the use of a low power laser beam, the beam patterns with concentric ring-shaped structure like Fig.2(d) are formed when the focal point positions are located in the sample cell. For a high concentration solution and with the use of a high power laser beam, the beam patterns with rich structure like Fig.2(e) are formed directly, and no pattern like Fig.2(d) is observed. With the use of an unfocused laser beam, the transmitted beam is not found to widen. For the focal point position located before and behind the sample cell, the

images of the beam patterns taken are shown in Fig.3. Obviously, the spot centre is bright when the focal point positions are located before and in the sample cell; in contrast, the spot centre is dark when the focal point positions are located behind the sample cell. With the changes of the focal point position, the laser power density irradiating the sample also changes. It is found that the incubation time of the transmitted beam widening is inversely proportional to the laser power density in the sample.



(a) (b) (c)

Fig.3. Images of the beam patterns on the screen for the focal point positions located (a) before, (b) in, and (c) behind the sample cell.

4. Discussion and analysis

With the theory of the optical-field-induced refractive index changes and that of Fresnel–Kirchhoff diffraction integrals,[8,15,16] the intensity distributions of the transmitted beams can be calculated when the focal point positions are located before and in, as well as behind the sample cell. The calculated intensity distributions for media with the positive refractive index increment with the increase of the laser power density are similar to the intensity distributions shown in Fig.3. Therefore, we think that the pattern formation in homogeneous polymer solutions induced by a CW laser can be explained by optical-field-induced refractive index changes.

We measure the optical absorbed spectrum of PI (shown in Fig.4) with Shimadzu UV–VIS–NIR Scanning Spectrophotometer (3101PC) in the experiment. The absorbed coefficients of PI in the range of the visible spectrum are small while they are large in the range of UV spectrum. Thus the possibilities of the refractive index changes caused by photothermal and multiphoton absorbtion effects can be excluded.[14] And the CW laser power density used in the experiment is so small that the particle trapping effect can not occur, so the possibilities of the refractive index changes caused by the particle trapping effect can also be excluded.[14] Considering that light-induced polymerization in the solutions of 1,4-PI in toluene can produce some products which are hard to dissolve in the solutions,[17] we think the light-induced polymerization may be responsible for the refractive index change and memory effect in the solutions. And the cooperation between C_{60} with PI in the solution[18] can make a contribution to the refractive index changes.

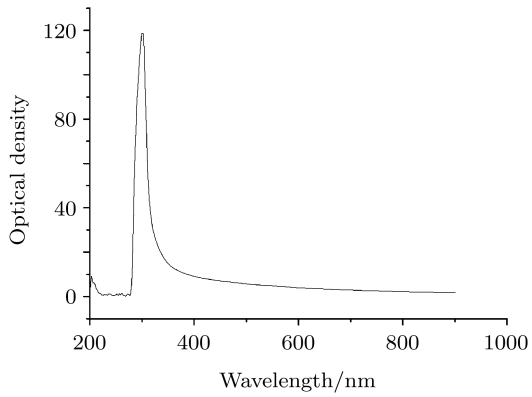


Fig.4. Optical absorbed spectrum of polyisoprene.

5. Conclusions

The pattern formation in homogeneous solutions of 1,4-PI in toluene saturated with C_{60} induced by a CW visible laser beam is observed in the experiment. The transmitted beam patterns change with the increase of the laser irradiation time. In the initial phase, the patterns with concentric ring-shaped structure are formed. In the end, the patterns become speckle-shaped. The transmitted spot centre is bright when the focal point positions are located before and in the sample cell; in contrast, the transmitted spot

centre is dark when the focal point positions are located behind the sample cell. The incubation time of the transmitted beam widening is inversely proportional to the laser power density and solution concentration. The pattern formation in homogeneous polymer solutions induced by a CW laser can be explained by optical-field-induced refractive index changes, but the mechanism of optical-field-induced refractive index changes in the polymer solutions needs to be further studied.

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