Nonlinear Molecular Reorientation of Polymer-Stabilized Dye-Doped Liquid Crystals by Depolarized Laser Beam

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Liquid crystals (LCs) are the innovative materials that can control their molecular orientation using an optical field with a laser beam. However, the molecular orientation has been explored only by a polarized laser beam. In this study, we report the molecular reorientation of oligothiophene-doped LCs by an irradiation with a focused depolarized laser beam that has a spatial polarization pattern. Two self-diffraction rings occurred by nonlinear molecular reorientation induced by the irradiation with the focused depolarized laser beam. The threshold intensities measured by the focused depolarized laser beam were two times higher than that of the linearly polarized laser beam.

Keywords: Liquid crystals, Nonlinear optical effect, Depolarized laser beam

1. Introduction

Liquid crystals (LCs) have drawn much attention for a fabrication of innovative materials and devices [1-3] applied to LC displays [4], tunable optical metamaterials [5], smart windows [6] and reconfigurable optical elements [7], as molecular reorientation is able to modulate the optical properties. In particular, the photoinduced orientational changes [8-11] enable one to develop all-optical devices including data storage [12], efficient optical switching [13] and light beam modulation [14]. The reorientation of LC molecules in optical field is enhanced by adding a small amount of absorbing dye molecules, which generally causes a photochemical reaction, to the LCs [15-24]. On the other hand, molecular reorientation without a photochemical reaction, termed photophysical process, has also been explored [25-29] because of exhibiting a well-defined threshold intensity of nonlinear molecular reorientation. We found that homeotropic-aligned LCs doped with an oligothiophene-dye (TR5) efficiently underwent the photophysical molecular reorientation with lower light intensity compared with previously reported dye-doped LCs [30]. In addition, polymer-stabilized dye-doped LCs (PSLCs) which were stabilized with a low concentration of LC polymers enabled us to change molecular orientation with a low threshold intensity [31]. Although the threshold intensity has been successfully reduced, the molecular reorientation behavior in the photophysical process was induced only by a polarized laser beam. In order to expand the versatility to the all-optical applications, molecular reorientation induced by not only a polarized beam but also a depolarized beam is desired.

In this paper, we report that the molecular reorientation of an oligothiophene-doped PSLC can be induced by a depolarized laser beam that has a polarization pattern. Two self-diffraction rings appeared by nonlinear molecular reorientation because a depolarized laser beam was split into two beams by focusing. The threshold intensities measured by a focused depolarized laser beam were two times higher than that of the linearly polarized laser beam.
2. Experimental

A host nematic LC, 5CB (4-cyano-4’-pentyl biphenyl), was obtained from Merck Ltd., Tokyo, Japan. A4CB (4-[4-(4’-cyanobiphenyl)oxy]butyl acrylate) was mixed to host LC as a monoacrylate according to a previous report (A4CB:5CB = 10:90, molar ratio) [29]. The dye molecule, TR5 (5,5’-Bis-(5-butyl-2-thienylethynyl)-2,2’:5’,2’’-ter thiophene) was synthesized as reported previously [30] and doped into the mixture of 5CB and A4CB at the concentration of 0.1 mol%. Irgacure 651 was used as the photoinitiator. After the prepared mixture was dissolved in THF and stirred for 1 h, the solvent was completely removed under vacuum condition. The mixture was inserted into a 100 μm thick glass cell treated with a silane coupler that provided the homeotropic alignment. The sample was irradiated with UV light (1.0 mW/cm², 365 nm, 1 h) from a high-pressure mercury lamp equipped with neutral density filters and band-pass filters (ND-70, UV- D36A, UV-35, IRA-25S, AGC Techno Glass, Co., Ltd., Shizuoka, Japan) for photopolymerization, followed by an annealing process.

The initial molecular orientation of the obtained PSLC and the dye in the sample were confirmed by a conoscopic image and polarized ultraviolet (UV)–visible (vis) absorption spectra. The conoscopic image and spectra were measured with a polarized optical microscope (BX50, Olympus, Tokyo, Japan) and polarized UV-vis absorption spectrophotometer (V-650ST, JASCO Corporation, Tokyo, Japan), respectively.

The photoinduced molecular reorientation of the TR5-doped PSLC was induced by the irradiation with a pump laser beam. The optical setup used in this study is shown in Fig. 1. A linearly polarized diode pumped solid state (DPSS) laser beam with a wavelength of 488 nm was used as a pump laser beam. The beam was modulated by a depolarizer (Thorlabs, Inc. DPP25-A) that has spatial grating retardation pattern. The shape of a depolarized laser beam was the same as that of an incident linearly polarized laser beam, while the polarization direction of the depolarized beam was periodically changed unlike that of the linearly polarized beam as shown in Fig. 2a. The depolarized beam was focused by a biconvex lens, L4, and subsequently the diffractive rings generated on the screen were analyzed by comparison with the rings by a linearly polarized laser beam. Upon exposing the LC cell to a pump beam with sufficiently high intensity, diffraction ring pattern appeared on a screen placed behind the sample arising from self-phase modulation induced by homeotropic-to-planar molecular reorientation. The light intensity, at which the diffraction ring was firstly observed with a beam profiler, was defined as the threshold of molecular reorientation. We compared the diffraction ring formation behavior and the threshold intensities of depolarized and linearly polarized laser beams.

3. Results and discussion

The prepared sample showed a uniform and optically transparent yellow color over the area. A conoscopic image of the polarized optical
microscope exhibited clear isogyre. In addition, the vertical and horizontal polarized UV–vis absorption spectra became coincident in 350-600 nm which corresponds to the absorbing bands of the host LCs and the guest dyes. These results show that the dye and LC molecules in the cell were homeotropically aligned.

When the homeotropic-aligned PSLC cell was irradiated with a focused depolarized laser beam, two diffraction rings were observed on the screen above a certain light intensity as shown in Fig. 3. When the light intensity was 5.0 W/cm², no diffraction ring pattern was shown. Above the threshold intensity, two diffraction rings appeared independently. The number of rings increased with the light intensity in the same manner, and finally reached 29 and 27 at 40.7 W/cm² for left and right rings, respectively. It means that the PSLC enables one to induce molecular reorientation even using the depolarized laser beam. Beam profile observation at the focal point revealed that the depolarized laser beam was split into two beams (Fig. 2b). Peak top intensities of the two beams were almost equal. The beam diameters were approximately 50 µm which were the same as that of a linearly polarized laser beam. The distance between the two beams was 50 µm, which is caused by diffraction of a focused depolarized laser beam.

Fig. 3. Typical diffraction ring patterns formed on a screen induced by a focused depolarized beam through a PSLC cell at various light intensities.

The number of right and left diffraction rings as a function of incident light intensity of focused depolarized laser beams are shown in Fig. 4. It is assumed that the light intensities of the focused depolarized laser beams are equally split into two. In each case, self-diffraction rings were observed above certain light intensities and the number of rings increased in accordance with light intensity. To investigate the photoresponsive behavior, the threshold intensity was defined as the light intensity at which the first diffraction ring appeared, and then measured with a beam profiler. As a result, the threshold intensities of right and left rings with the focused depolarized laser beams were 8.32 W/cm² and 7.63 W/cm², respectively. These intensities were two times higher than that of the linearly polarized laser beam (3.30 W/cm²). The threshold intensity is determined by balance of torques: the optical-electric torque, the external electric torque, bulk elasticity, and surface anchoring. Among them, the optical-electric torque is generated by the interaction between the optical field and the dye molecule, that is, a torque direction is parallel to the polarization direction. On the other hand, the polarization direction of the focused depolarized laser beam was random compared with a linearly polarized laser beam. Such polarization direction induces the random optical-electric torque direction, resulting in the increase of the threshold intensity. We revealed that nonlinear molecular reorientation of oligothiophene-doped PSLC occurred even with the depolarized laser beam.

![Depolarized and linearly polarized laser beams](image-url)

Fig. 4. Number of diffraction rings induced with depolarized (blue and red dots) and linearly polarized (gray dots) beams as a function of light intensity.

4. Conclusion

In conclusion, nonlinear molecular reorientation of an oligothiophene-doped PSLC occurred even by the irradiation with a focused depolarized laser beam. The depolarized laser beam was split into two beams at the focal point. Two diffraction rings appeared on the screen, and the number of rings
increased as the light intensity was increased. The threshold intensities of the focused depolarized laser beams were approximately two times higher than that of the linearly polarized laser beam, which may be due to the randomized polarization of the focused depolarized laser beams at the focal point. We believe that the nonlinear molecular response of the dye-doped LCs enables much wider applications of optics.

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