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Polarization Recording in Photoinduced Chiral Material for Optical Storage

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A novel method of optical data storage was proposed using the rotation of polarization. The optical data was recorded as a chiral structure formed in a film made of an azobenzene copolymer using elliptically polarized light irradiation. The elliptically polarized light of a reading light was digitized into two states, "0" and "1". The initial state is defined as "0", while state "1" is observed by an optical rotation of the reading light, which is achieved by the photoinduced chiral structure formation. The recording characteristics were investigated by varying the intensity of the recording light and the recording time. Since the chiral structure was erased by circularly polarized light, state "1" could be reversed to state "0". The possibility of using our proposed method for achieving next-generation rewritable, multilevel, and parallel optical data storage was discussed. [DOI: 10.1143/JJAP.46.3928]

KEYWORDS: optical data storage, azobenzene, photoinduced birefringence, photoinduced chirality, optical rotation

1. Introduction

During the last decade, various unique phenomena have been observed in photoresponsive azobenzene materials such as photoinduced birefringence,[1,2] photoinduced surface relief formation,[3,4] photoinduced chirality,[5,6] and photodriven actuation.[7] All these originate from the photoisomerization characteristics of the azobenzene materials. These phenomena are potentially useful for the development of novel active optical devices, photodriven mechanical devices, and optical data storage. In this paper, a novel optical recording method is proposed on the basis of the photoinduced chirality of an azobenzene copolymer reported in our previous papers.[8–12]

It is well known that when linearly polarized light is irradiated on an azobenzene material, the molecules are aligned in the direction perpendicular to the polarization axis. Then, birefringence is induced in the azobenzene material. However, the induction of optical birefringence is not limited to linearly polarized light irradiation, it is also possible in elliptically polarized light (EPL) irradiation. When left-handed elliptically polarized light (L-EPL) is irradiated on an azobenzene film, the polarization is spontaneously rotated clockwise. In contrast, in the case of right-handed elliptically polarized light (R-EPL), the polarization is rotated counterclockwise. The spontaneous rotation of polarization is observed even if the azobenzene material is initially amorphous and achiral. The rotation of polarization can be explained by the fact that chirality is induced in azobenzene material. From these characteristics, the photoinduced chirality has a potential for optical data storage similar to that of a magneto-optical (MO) disc. In addition, our proposed method may lead to the parallel and multilevel recording of polarization, whereas the optical data is recorded by a bit-by-bit recording in the MO disc. In this paper, the possibility of polarization recording is investigated for a next-generation optical data storage method.

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2. Theory

2.1 Photoinduced chirality

Photoinduced chirality is based on the molecular reorientation in azobenzene material triggered by *trans–cis–trans* photoisomerization cycles. When *trans*-azobenzene absorbs light, the conformation is changed to *cis*-form. The *cis*-azobenzene immediately relaxes back to the *trans*-form in thermal processes. The direction of *trans*-azobenzene that has relaxed back from the *cis*-form is not always the same as the original one. However, once the *trans*-azobenzene molecules are aligned perpendicularly to the axis of linearly polarized light, they cannot absorb the light any more, and the direction of the molecules is maintained for a while. As a result, molecular reorientation occurs and birefringence is generated. Under the irradiation of EPL, the molecules are aligned in the direction perpendicular to the principal axis of the EPL and birefringence is also generated. Then, the polarization state is changed by the interaction between the photoinduced birefringence and the EPL, because not only the refractive index but also the absorption coefficient becomes anisotropic. Therefore, the polarization state is changed.

However, it is often pointed out that the molecular orientation of conventional azobenzene is easily relaxed and that birefringence is also reduced by the effect of a thermal disturbance under ambient conditions. Thus, we have developed a new azobenzene copolymer with good stability of the phototriggered molecular alignment. The copolymer consists of the 2-[4-(4-cyanophenylazo)phenoxy]ethyl moiety and the 2-[2-methyl-4-(4-phenylazophenylazo)phenoxy]-ethyl moiety at a 50 : 50 molar ratio (PCDY50). Its chemical structure is shown in Fig. 1. The left part of this copolymer acts as a trigger of molecular reorientation. Also, the right part has a high molecular birefringence and its axis is cooperatively aligned in the direction parallel to the left part. The superior stability of the photoinduced molecular alignment may be attributed to the cooperative and mutual promotion of the phototriggered molecular reorientation.

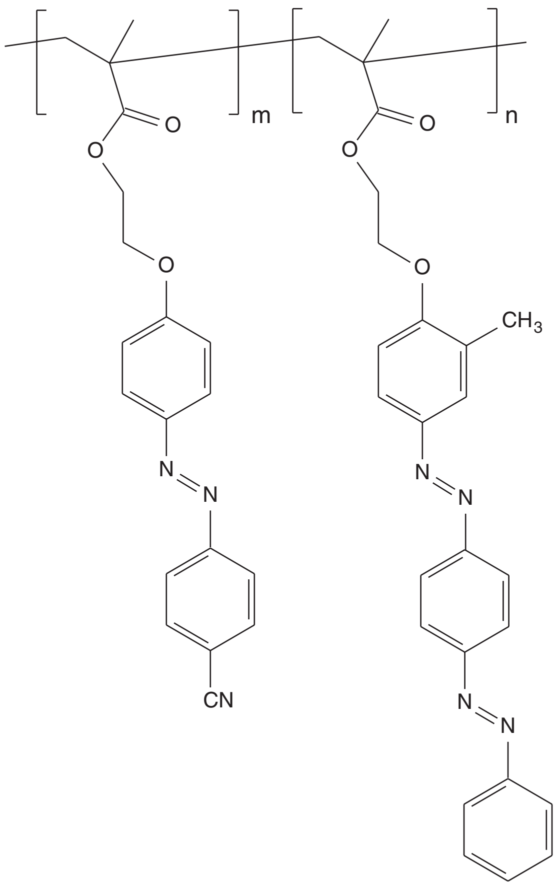


Fig. 1. Chemical structure of PCDY copolymer. The ratio of $m : n$ is 1 : 1 in PCDY50.

2.2 Polarization recording and reading

The optical data is recorded to an azobenzene film using an elliptically polarized laser beam of an appropriate wavelength. Then, chirality is induced in the film using the recording light, and the principal axis of an elliptically polarized reading light is rotated owing to the photoinduced chirality of the film. The reading light is recognized as having two states, namely "0" and "1" relative to its principal axis, as shown in Fig. 2. In practice, detection of the two states can be easily achieved by dividing the reading light into horizontally and vertically polarized light and both intensities are detected. The signal of the optical data s is defined as

$$s = \left\langle \frac{|E_x|^2 - |E_y|^2}{|E_x|^2 + |E_y|^2} \right\rangle, \tag{1}$$

where E_x and E_y are the horizontal and vertical components of the electric field, respectively. When the signal is positive $s > 0$, the state is defined as "0", as shown in Fig. 2(a). In contrast, in the case of $s < 0$, the state is defined as "1", as shown in Fig. 2(b). The rotation angle of polarization is expressed by an azimuthal angle α, which is the azimuth of the polarization ellipse with respect to the E_x axis. In the case of the binary coding, the boundary of the two states is set to 45° in azimuth. Namely, if the azimuthal angle is less than 45°, the horizontal component becomes larger than the vertical component and the state becomes "0". In contrast, the state becomes "1" when the azimuthal angle is larger than 45° Here, state "0" is assumed as the initial state. If R-EPL is irradiated on the azobenzene film, a chiral structure that induces counterclockwise optical rotation to the reading light is formed in the film. When the chiral structure is formed up to the point where the azimuthal angle becomes larger than 45° by the rotation of polarization, the vertical component becomes larger than the horizontal component and the state changes into "1". Unless the recording light is irradiated, the polarization state of the reading light with state "0" is not changed. It is already confirmed that the photoinduced birefringence can be erased by circularly polarized light irradiation. Therefore, state "1" can be reversed to "0" by light with circular polarization.

However, if we employ a laser beam whose wavelength is the same as the recording beam for data reading, the reading process is unavoidably destructive. In this case, significantly satisfactory durability cannot be expected for this data-reading method since the reading light does also affect the recording medium. For example, when we read state "0", the state may change to "1" because chirality is induced in the medium by EPL. In order to avoid the destructive reading, the intensity of reading light must be reduced so that the azimuth change is only nominally observed. Moreover, the use of a feedback system, as shown in Fig. 3, is effective for solving the problem. In this system, the polarization state of incident light is adjusted using a polarization modulator such as an electrooptic modulator, a magnetooptic modulator, and a photoelastic modulator. The optical data is recorded using EPL. In the reading process, the intensity of the EPL is reduced. Then, the reading light through the medium is divided into horizontally and

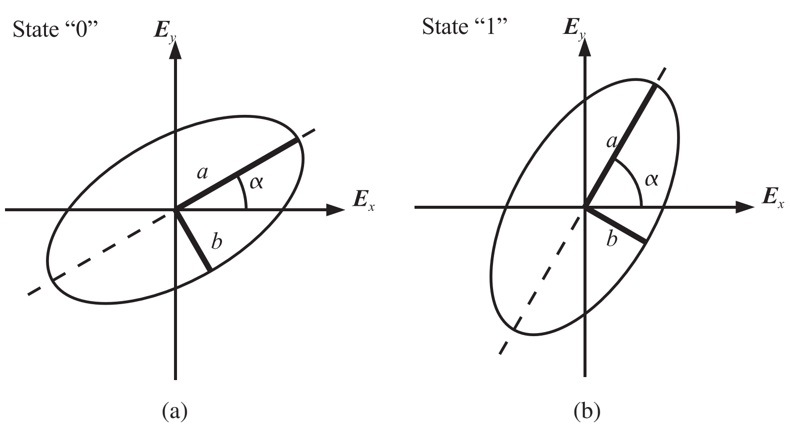


Fig. 2. States of elliptical polarization. (a) and (b) show the polarization ellipses of states "0" and "1", respectively. Here, E_x, E_y, a, b, and α are the x- and y-components of the electric field, and the long and short radii, and azimuthal angle of the polarization ellipse, respectively.

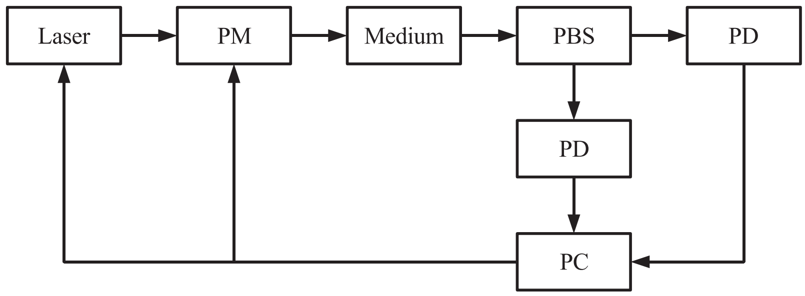


Fig. 3. Feedback system of polarization recording. PM, PBS, and PD are the polarization modulator, polarization beam splitter, and photodetector, respectively.

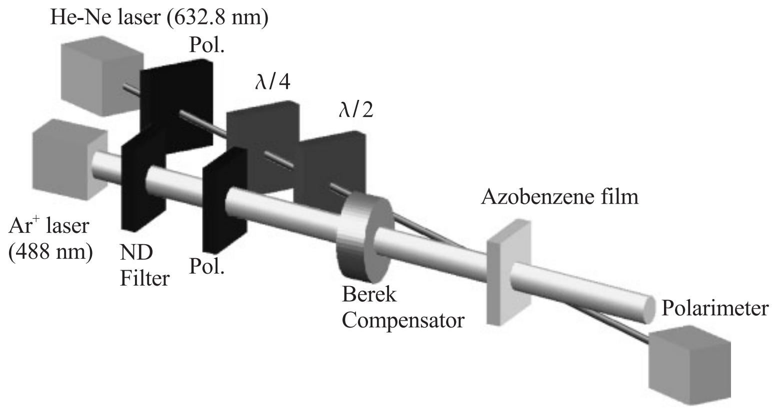


Fig. 4. Experimental setup of polarization recording.

vertically polarized light using a polarization beam splitter. The intensities of the two linearly polarized light beams are detected using two photodetectors, and the signal of the optical data is read. When the signal is detected as state "0", the polarization of the reading light is set to circular polarization in order not to change the state. Furthermore, the system is applicable for parallel recording using a spatial light modulator and charge-coupled devices as the polarization modulator and photodetectors. As another solution to this problem, a reading light with a wavelength that does not affect the recording medium can be used. Although the former method is practical, the latter method is used in this fundamental study, because the optical data recorded can be accurately read out and a feedback system is not necessary. The optical data can be read by a reading light with a different wavelength, because the rotation of polarization is brought about through a chiral structure formed by the recording light.

3. Experimental Procedure

3.1 Experimental setup

In this study, the PCDY50 copolymer, as shown in Fig. 1, was used as a recording medium. The synthesis method and characteristics of this azobenzene copolymer have been reported in our previous paper.[11] This azobenzene copolymer is achiral, and amorphous films with good optical quality can be obtained by the spin-coating method. The experimental setup of the polarization recording is shown in Fig. 4. The optical data was recorded in an azobenzene film using an Ar^+ laser of 488 nm. The irradiance of the recording laser beam was adjusted using a ND filter. The diameter of the recording area was 3 mm. A He–Ne laser with a wavelength of 632.8 nm was used as a reading light. The polarization states were controlled using polarizers and a compensator or waveplates. The ellipticities of the recording and reading lights were set to +0.5 and +0.8, respectively. In order to read the optical data, the Stokes parameters of the reading light were measured using a polarimeter.

3.2 Signal processing

The Stokes parameters are represented by

$$S_0 = \langle |E_x|^2 + |E_y|^2 \rangle, \tag{2}$$

$$S_1 = \langle |E_x|^2 - |E_y|^2 \rangle, \tag{3}$$

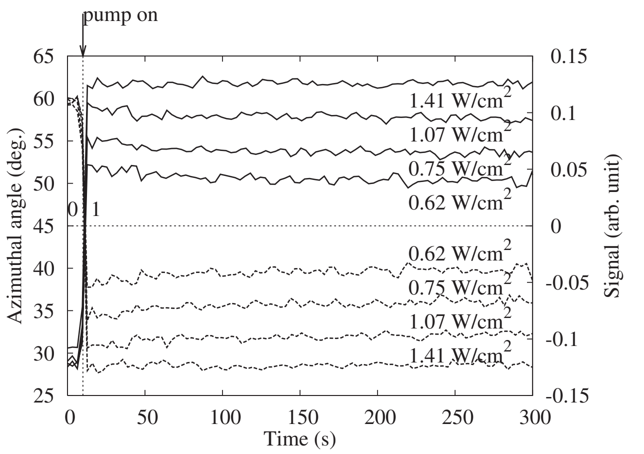
$$S_2 = \langle E_x E_y^* + E_x^* E_y \rangle, \tag{4}$$

$$S_3 = \langle \mathrm{i}(E_x E_y^* - E_x^* E_y) \rangle. \tag{5}$$

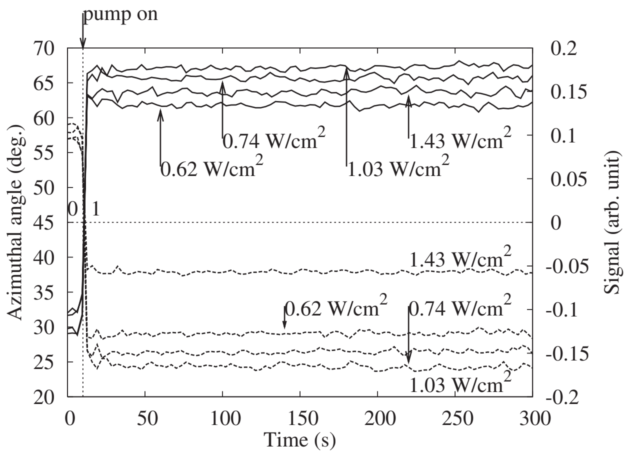
The signal s can be obtained as

$$s = \frac{S_1}{S_0}. \tag{6}$$

In this paper, the azimuthal angles of the recording and reading lights were set horizontally in order to recognize the angle of 0° in the polarimeter. When the angles are close to 45°, it is easy to change the state of the signal because the azimuthal angles cross the boundary with a small change. The azimuthal angles used in practical data analysis were transformed to desired values using Stokes parameters measured using the polarimeter and Mueller matrices. Conversely, this procedure means that the boundary angle of 45° is changed. Although the actual angles of the reading EPL can be varied using a half waveplate, experimental errors caused by the mechanical rotation of the waveplate may occur. These anticipated errors can be excluded by the following analytical procedure. The azimuthal angles determined by Mueller matrices can be obtained as



(a)



(b)

Fig. 5. Light intensity dependence of polarization recording. The recording was started at 10 s. Solid and dashed lines show the time evolutions of the azimuthal angle and the signal, respectively. (a) and (b) show the experimental results in the cases of the recording durations of 1 and 2 s, respectively.

$$\begin{bmatrix} S'_0 \\ S'_1 \\ S'_2 \\ S'_3 \end{bmatrix} = \begin{bmatrix} 1 & 0 & 0 & 0 \\ 0 & \cos(2\alpha') & -\sin(2\alpha') & 0 \\ 0 & \sin(2\alpha') & \cos(2\alpha') & 0 \\ 0 & 0 & 0 & 1 \end{bmatrix} \begin{bmatrix} S_0 \\ S_1 \\ S_2 \\ S_3 \end{bmatrix}, \quad (7)$$

where (S'_0, S'_1, S'_2, S'_3) are the Stokes parameters with the azimuthal angle of α'. The signal after azimuth change is expressed by

$$s' = \frac{S_1 \cos(2\alpha') - S_2 \sin(2\alpha')}{S_0}. \quad (8)$$

The sign of s' was used to evaluate the state of the binary code.

4. Results and Discussion

The polarization recording efficiency is dependent on the intensity of the recording light and the recording duration. The dependence on light intensity is shown in Fig. 5. The fluctuation of the signal is not due to the sample or the excitation beam quality but simply due to the electric noise of the photodetector. Figure 5(a) shows the experimental results when the recording light was irradiated for 1 s. The recording was started at 10 s and the signals were measured for 5 minutes after the recording is stopped. The light intensity was varied from 0.6 to 1.4 W/cm^2 and the azimuthal angle was set to 30° As shown in Fig. 5(a), optical rotations and signal changes were observed under the recording, and it was confirmed that state "0" is changed into "1". The optical rotations and signal changes were maintained for at least 5 minutes after the recording. When the intensity of the recording light was large, the rotation angle and the contrast of the signal became large under this condition. In the case of a recording duration of 2 s, the rotation angle and the contrast of the signal were reduced, as shown in Fig. 5(b). From these results, it was found that there is an optimal relationship between the light intensity and the recording duration.

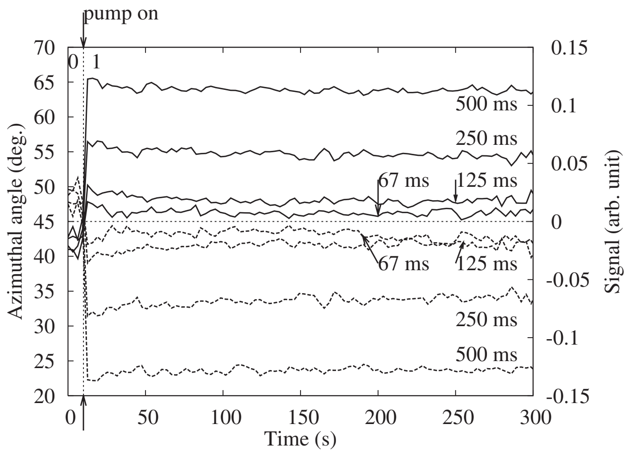


Fig. 6. Recording duration dependence. The recording was started at 10 s. Solid and dashed lines show time evolutions of the azimuthal angle and the signal, respectively.

According to the intensity dependence, a recording light intensity of 1.4 W/cm^2 is not too large to reduce the signal contrast in the case of a recording duration for cases shorter than 1 s. Therefore, the recording duration dependence shorter than 1 s was investigated using the recording light with the intensity of 1.4 W/cm^2. Figure 6 shows the experimental results. In this experiment, the azimuths of the recording and reading light polarizations were set to 41°. When the recording duration becomes short, the rotation angles and signal contrasts naturally decrease. However, note that the recording could be performed even if the recording duration was shorter than 100 ms under these conditions. In this paper, the 67 ms recording was achieved using the recording light of 1.4 W/cm^2. Of course, it is true that our study is still primitive and that the experimental conditions for the recording and reading lights are also not yet optimized. However, we believe that the recording duration can be reduced much more via further study of the recording system and additional material development.

State "1" recorded using EPL can be reversed to state "0" by illumination using circularly polarized light. In this experiment, right-handed circularly polarized light is used to reverse state "1" to state "0". The intensity of the circularly polarized light is set to 1.4 W/cm^2, which is the same as that of the R-EPL used for recording state "1". Figure 7 shows the experimental results of the reversibility. State "1" was recorded by illumination using the recording light for 1 s [cases (a)–(c)] or 250 ms [case (d)] at 10 s. Then, the

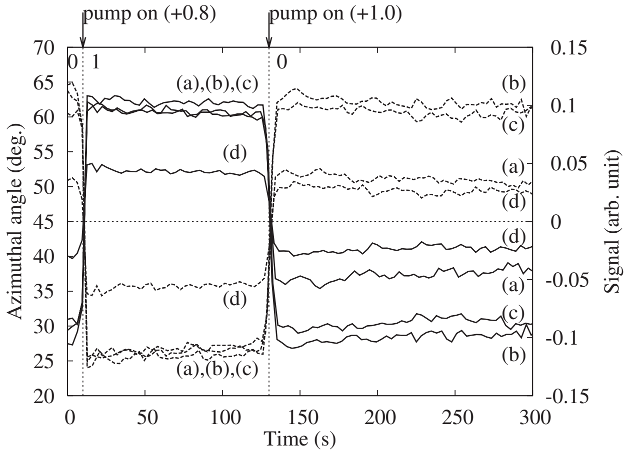


Fig. 7. Reversible characteristics of polarization recording. The recording of state "1" was started at 10 s and the state was reversed to "0" at 130 s. Solid and dashed lines show the time evolutions of the azimuthal angle and the signal, respectively. In the cases of lines (a), (b), and (c), the recording duration at 10 s was set to 1 s and the input azimuthal angle was set to 30°. In the case of line (d), the recording duration was set to 250 ms and the input azimuthal angle was set to 40°. The recording durations at 130 s of (a), (b), (c), and (d) were 1, 2, 3, and 1 s, respectively.

azimuthal angles of recording and reading lights were set to 30° [cases (a)–(c)] or 40° [case (d)]. State "1" was reversed to state "0" within 2–3 s by the circularly polarized light at 130 s. The signals of state "1" recorded for 1 s were almost reversed to the initial state when the duration of the reversal was longer than 2 s, as shown in Figs. 7(b) and 7(c), although the signal was not reversed in the case when the duration of the reversal was 1 s, as shown in Fig. 7(a). When the the recording duration was shorter than 1 s, as in the 250 ms recording, the signal could be almost reversed to the initial state by the 1 s irradiation of the light used to reverse the state, as shown in Fig. 7(d). It was found that a longer time than that required to reach state "1" during the recording was necessary for reversal to state "0". The rewritable optical recording was demonstrated for 100 ms–2 s. The recording and reversing times can be reduced by employing more intense light.

In our proposed method, the ellipticity change in the polarization ellipse is less than 5% for all optical rotation angles. The border of signal recognization can be set to any angle in the range from 0 to 90° and the number of borders is not limited to one, i.e., if we assume that we set the number of borders to more than one at appropriate rotation angles, multilevel data recording can be achieved using the same system as that described in this paper. Such a multiple data coding technique is promising for improving the recording capacity. In addition, parallel recording is possible because the optical data can be recorded in the area of a 3-mm-diameter circle with the optical power of 100 mW, which is the general recording power used for present optical discs. For example, when the recording data is recorded as a spot of 3 µm diameter, it is estimated that 10^6 bits can be recorded in parallel at the recording power of 100 mW. The polarization states of the 10^6 bits of data can be modulated in parallel using an SLM. The spot size of 3 µm diameter and 10^6 pixels on the SLM can be realized because the spot size is about ten times larger than the diffraction limit of visible light and a one megapixel SLM is commercially available.

In addition, these recording and reading methods can be achieved using linearly polarized light, because the rotation of polarization is also observed in general birefringent films (such as a half waveplate) when the polarization axis of the reading light is different from the birefringent axis. If we use the azobenzene film as a recording medium, linear birefringence can be brought about by the irradiation of linearly polarized light. Thus, we can perform the recording and reading processes on azobenzene thin film using only the linearly polarized light. However, another polarization modulator is required to change the polarization axis for recording and reading in this case. Moreover, if we assume multilevel recording by changing the azimuthal angle of the reading light, we need many boundaries in the azimuth of the reading light. In the case of a linear birefringent medium, the azimuthal angle of the reading light oscillates around the axis of birefringence with respect to the phase difference between the horizontally and vertically polarized light. The range of the change in azimuth and the number of boundaries are limited because the rotation direction of polarization is reversible. Therefore, recording and reading methods using linearly polarized light are not appropriate for multilevel recording.

5. Conclusions

In this paper, a novel method of optical data storage was proposed using photoinduced chirality in azobenzene copolymer films. The chiral structure is formed in the film by illumination using EPL. When a reading light passes through the chiral strucure, the principal axis of EPL is rotated. In other words, the optical data is identified by the azimuthal angle of the principal axis. In our proposed method, these characteristics were used for optical data storage. The signal change was dependent on the intensity of the recording light and recording duration. In this study, 67 ms recording was achieved using a recording light of 1.4 W/cm^2. The recording time can be reduced using more intense light. The reversibility of the optical data recorded in the film was also demonstrated. From this fundamental study, the possibility of rewritable, multilevel, and parallel optical data storage using this method was indicated.

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