All-Optical Polarization Control Using a Nonlinear Optical Medium with Chirality.

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We have proposed two schemes of all-optical polarization control with optical nonlinearity of chiral media. One is the nonlinear polarization change caused by circular anisotropy of the nonlinear refractive index. The other is the nondegenerate scheme of all-optical polarization control by combination of linear optical rotatory dispersion and nonlinear refractive index. Both methods underlie an optical Kerr shutter which is operable by colinearly polarized two beams. We demonstrate optical Kerr shutter and optical XOR using a solution of chiral materials. We also show the device structure could be very simple, and show the idea of devices which could be called all-optical spatial processing plate.

1.INTRODUCTION

Conventional Optical Kerr shutter(OKS) system has three directional polarization beams, vertical probe, 45° pump and horizontal signal beam. Intensity dependent polarization self rotation must use the elliptically polarized beam¹). Chiral material has circular anisotropy (optical activity) which causes the optical rotatory power and its wavelength dispersion(ORD). We have researched different two schemes which are all-optical polarization control with colinearly (same directional linearly) polarized lights by combination of the chirality and the nonlinear refractive index. If OKS were operable with colinearly polarized pump and probe beam, device structure could be made very simple. First scheme is using the circular anisotropy of the nonlinear refractive index (Chiral nonlinearity)²⁾. Second is a nondegenerate scheme with the combination of ORD and nonlinear refractive index(ORD-NLO)³⁾. In this paper we report the principles and the demonstration results of the OKS using the solution of chiral materials about each We also discuss method. the device applications.

2. CHIRAL NONLINEARITY

A chiral molecule has the optical isomer. We have (+), (-) media at the chiral media, and also have racemic media(\pm) which contains the equal amount of (+) and (-) molecules. The optical rotational angle is,

 $\eta = (\pi 1 / \lambda) (\mathbf{n}_{\mathrm{L}} - \mathbf{n}_{\mathrm{R}}). \qquad (2.1)$

where l is path length , λ is wavelength and nL and nR are the refractive indices for left (L)

and right (R) polarization component. (+) and (-) media have optical activity, and (-) molecule is the mirror image of (+) molecule. Optical activity is kept even in a medium composed with random directional molecules such as liquid or doped polymer. The relationship of refractive indices are,

 $n_{L}^{(+)} = n_{R}^{(.)} \neq n_{R}^{(+)} = n_{L}^{(.)}$ (2.2)

and,

 $\eta (+) = -\eta (\cdot) \tag{2.3}$

therefore, racemic media are optical inactive. When linearly polarized intense beam

irradiate the sample, refractive indices are changed, and magnitude of the change for L and R polarization is different. Then nonlinear rotational angle is

$$\delta \eta_{nl} = (\pi 1 / \lambda) n_{2chi}I_{pp}$$
, (2.4)
where $n_{2chi} \equiv (n_{2L} - n_{2R})$, n_2 is nonlinear
refractive index, subscript, L, R means for L
and R polarized light respectively, I_{pp} is pump
intensity. In optical active media $n_{2L} \neq n_{2R}$ and
OKS is operable with colinearly polarized
beams, and also intensity-dependent-self-
polarization-rotation can be occurred by
linearly polarized light.

We have demonstrated OKS with colinearly polarized beams using the solution of some helicenes. In this paper we show the experimental result using the 0.05wt/vol% (5mg/10ml) THF solution of thiaundecahelicene (TH[11]) at the resonant region. The solution of (+), (-) isomer or (\pm) mixture is used with 2mm path. To avoid an ellipticalization of probe beam and to detect the polarization change sensitively, we set the

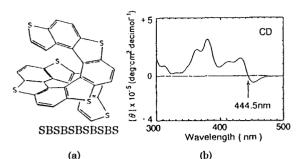


Fig.1 Illustration of structure (a) and CD spectrum (b) of (+)-TH[11].

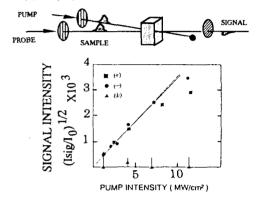


Fig.2 Pump intensity dependence of Isig of THF solution of TH[11]s.

wavelength 444.5nm at which CD spectrum cross to zero.

Detail of experimental setup is the same as that reported in ref.2 except for wavelength. Illustration of colinear OKS is shown in fig.2. The signal intensity I_{sig} is expressed with nonlinear rotation angle $\delta \eta_{nl}$ as,

 $(I_{sig}/I_0)^{1/2} = |\sin| \delta \eta_{nl}||$. (2.5) where I_0 is the total intensity of the transmittable probe beam. Fig.2 shows the result of pump intensity dependence of signal intensity. OKS signal is clearly observed in (+) and (-) media. (but signal is not observed in recemic media.) This is the evidence of chiral nonlinearlity. The nonlinear refractive indices effective for chiral nonlinearity n_{2chi} is the order of 10^{-14} (cm²/W) ($\alpha = 12$ cm⁻¹). We also examine some more helicenes, but all α normalized n_{2chi} are the order of 10^{-15} (cm³/W) which containes at largest few percent of OKS effective n_2 .

3.ORD-NLO

Optical rotatory power has a wavelength

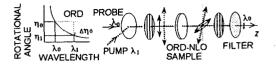


Fig.3 Schematic diagram of ORD Kerr shutter.

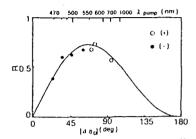


Fig.4 | $\Delta \eta_0$ | dependence of R of 4% optically active [6] solution in CS₂. Solid curve is calculated value. Probe wavelength is fixed at 450nm.

dispersion. When colinearly polarized two beams each having a different wavelength are introduced into optical active medium, the angle θ between two polarization planes is generated while both beams are traveling through the medium. If this medium has a nonlinear refractive index, OKS could be operated. Schematic diagram of the ORD-Kerr shutter is shown in fig.3. In this case pumping angle θ is in propotional to z position (z=0 at the entrance of sample). Then perpendicular component intensity of probe beam I_{sig} can be expressed using OKS effective n₂ as (see ref.3),

 $[(I_{sig}/I_0)^{1/2}/I_{pp}]_{ORD} = \pi 1 / \lambda_0 \cdot R |n_2|, (3.1)$ where

 $\mathbf{R} = |(1 - \cos 2 \,\Delta \eta_{o}) / 2 \,\Delta \eta_{o}|, \qquad (3.2)$

where $\Delta \eta_{o} = \eta_{\text{probe}} - \eta_{\text{pump}}$ is the difference of angles at the exit. Let's compare eq.(3.1) with the equation of 45° OKS.

 $[(I_{sig}/I_o)^{1/2}/I_{pp}]_{45^\circ} = \pi 1/\lambda_{0^\circ} |n_2|. \quad (3.3)$ The eq.(3.1) means ORD-OKS utilizes $|n_2|$ with the efficiency of R.

Detail of experimental setup is reported in ref.3. We use CS₂ solutions of hexahelicene ([6]) of 4wt/vol% with a path of 2cm as the sample. Solution of optical active (+),(-)-[6] is used for ORD-OKS, optical inactive (\pm)-[6] is used for 45° OKS for comparison. We measure the pump intensity dependences of I_{sig} on

ORD-OKS and 45° OKS.

ORD-OKS signal is clearly observed, and we determine the R using eqs.(3.1) and (3.3). Probe wavelength is fixed at 450nm, then $\Delta \eta \circ$ can be determined from pump wavelength

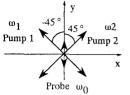


Fig.5 Definition of axes.

(see fig.3). We measure the $|\Delta \eta_{\circ}|$ dependence for R varying the pump wavelength. Results is shown in fig.4. The solid curve shows calculate value. Theoretical analysis show the maximum R value of 0.72 at $|\Delta \eta_{\circ}| = 66.5^{\circ}$, and the experimental result shows good agreement with calculated value. After all, the ORD-OKS is comparable to the conventional 45° OKS having 0.72 times of the $|\eta_2|$ value.

4. APPLICATIONS OF DEVICE 4.1 ORD-XOR

OKS is useful not only simple switching device various all-optical but signal processing. For example G.Jonusauskas et al. demonstrated a very fast OKS in which Kerr signal was canceled by delayed pump beam⁴⁾. OKS with the plural pump beams enable the all-optical operation. If we set the threshold correctly, logical operation could be realized. Before the subject, let's consider the OKS with two pump beams which have $+45^{\circ}$ and -45° polarization from probe polarization respectively (fig.5). Then x and y components of the electric field are

 $\mathbf{E}_{\mathbf{x}} = 1/2 (\varepsilon_2 \mathrm{e}^{\mathrm{i}\omega_2 \mathrm{t}} \mathrm{sin} 45^\circ - \varepsilon_1 \mathrm{e}^{\mathrm{i}\omega_1 \mathrm{t}} \mathrm{sin} 45^\circ + \mathrm{c.c.}),$

E_y=1/2(
$$\varepsilon_0 e^{-i\omega_0 t} + \varepsilon_2 e^{-i\omega_2 t} cos 45^\circ$$

+
$$\varepsilon_1 e^{i\omega_1 t} \cos 45^\circ$$
 +c.c.).

where subscripts 0,1,2 mean for probe, pump1 and pump2 respectively. Then x component of nonlinear electrical polarization at the nonresonant region in isotropic medium is

$$P_{\mathbf{x}}(\omega_{0}) = 3/2 \left[\chi^{(3)}_{\mathbf{x}\mathbf{x}\mathbf{y}\mathbf{y}}(-\omega_{0};\omega_{2},-\omega_{2},\omega_{0}) \mid \varepsilon_{2} \mid^{2} - \chi^{(3)}_{\mathbf{x}\mathbf{x}\mathbf{y}\mathbf{y}}(-\omega_{0};\omega_{1},-\omega_{1},\omega_{0}) \mid \varepsilon_{1} \mid^{2} \right]$$

$$\times 1/2 \left(\varepsilon_{0} e^{-i^{\omega_{0}t}} + c.c. \right).$$
(3.2)

When we can ignore the wavelength dispersion of nonlinearity, perpendicular component intensity of probe beam is expressed by using pump intensities I_1 and I_2 as

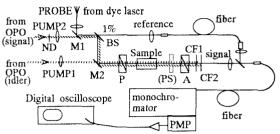


Fig.6 Experimental setup of XOR operation using ORD-OKS.P:polarizer,A:analyzer,F:color filter.

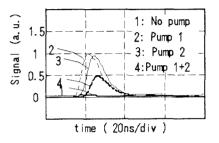


Fig.7 Signal of ORD-XOR output. 1-4 are shifted respective 2ns to avoid to overlap the waveform

 $I_x(\omega_0) \propto (I_1 - I_2)^2$ (3.3)The output is the result of XOR operation between pump1 and pump2 on a condition of $I_1=I_2$. These polarization state can be realized with very simple set up by using the ORD-NLO. Where pump 1,2 and probe wavelength are selected as $\Delta \eta_1 = -\Delta \eta_2$, where $\Delta \eta_1 = \eta_1$ $-\eta_{0}\Delta\eta_{2}=\eta_{2}-\eta_{0}$. When linearly polarized three beams having same polarization are traveling through the medium, polarization of pump1 and 2 are to make angle from probe polarization, which are same magnitude and opposite sign. We can imagine the scheme of ORD-XOR by adding the one more pump beam to fig.3.

Experimental setup is shown in fig.6. We use for pump beams the signal (490nm) and idler (1288nm) beams of OPO laser (GWU BBO-OPO) pumped by THG of Nd:YAG laser (Lumonics HY400), and use for probe beam the dye laser (558nm). Pulse width is 7ns and repetition rate is 10Hz. Two pump beams and probe beam are overlapped by dichroic mirrors M1 and M2, and linearly polarized to same orientation by polarizer P. The beams are traveling through the sample, and pump

(3.1)

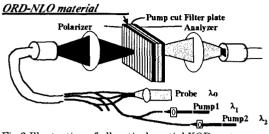


Fig.8 Illustration of all-optical spatial XOR system.

beams are cut by color filters F1 and F2. Probe beam is blocked by analyzer A under no pump beam. The signal beam passing through A is detected by photomultiplier PD1 (Hamamatsu R955) and digital oscilloscope (HP54502A). Sample is 3.45% CS₂ solution of (+)-[6] with path of 5cm. The rotational angle is $\eta_0 = 90^{\circ}$ at 558nm, is $\eta_2 = 158^{\circ} (\Delta \eta_2 = 68^{\circ})$ at 490nm, and η_1 is about 22° ($\Delta \eta_1 = -68^\circ$) at 1288nm. Output signals are shown in fig.7. Intensity of pump2 is adjusted by ND to make the same signl level on pumping with pump1. On the pumping of both pump1 and 2, signal level is decrease to less than half. This is considered to be XOR operation although it is imperfect. We confirm that this decrease is caused by polarization change on the measurement with elliptical polarization bias in probe beam. Then imperfection of XOR operation can be thought to be caused by bad profile of pump beams.

4.2 APPLICATION OF DEVICE

We have demonstrated OKS with the colinearly polarized beams. We show in this sub section why colinear scheme is advantageous by showing the idea of device applications. Fig.8 shows the illustration of the ORD-XOR system. Colinear polarization scheme make possible only one polarizer system for input beams. This means the parts from polarizer to analyzer can be united in one device, which could be called all-optical spatial processing plate. This structure cannot be realized without colinear polarization scheme. ORD-NLO is also possible to another applications. If this plate is introduced into laser cavity, logical

operating results will function as Q-switch. Then the device is added threshold and amplifier function to the basic operation. The spatial analogical subtraction device is obtained by adding the elliptical polarization bias to this plate. And optical FM-AM converter is obtained by using the frequency modulated pump beam, in which pumping angle is changed according to the frequency Thus these device modulation. are constructed with plate structure which could be called all-optical spatial processing plate. 5.CONCLUSION

We confirmed the OKS and XOR operation with colinearly polarized beams by using the nonlinear media having chirality. We showed the devices could be constructed with very simple plate structure by using colinear polarization scheme. But to realize these device, we need the more efficient material for nonlinearity and optical rotatory power. It is also subject that we study to induce the chirality in efficient nonlinear material.

Acknowledgments

The authors would like to gratefully acknowledge Prof. M.Kuwata-Gonokami for stimulating discussions and important advice.

This work was performed by UBE Industries Ltd. under the management of Japan High Polymer Center as a part of Industrial Science and Technology Frontier Program supported by New Energy and Industrial Technology Development Organization.

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