Enhancement of modulation depth of an all-optical switch using an azo dye-ethyl red film*

Lu Wen-Qiang(陆文强)[a], Chen Gui-Ying(陈桂英)[b]†, Hao Zhao-Feng(郝召锋)[c], Xu Jing-Jun(许京军)[c], Tian Jian-Guo(田建国)[c], and Zhang Chun-Ping(张春平)[c]

[a] *Physics Experimental Teaching Center, School of Physics, Nankai University, Tianjin* 300071, *China*
[b] *Key Laboratory of Tropical Marine Environmental Dynamics, South China Sea Institute of Oceanology, Chinese Academy of Sciences, Guangzhou* 510301, *China*
[c] *Photonic Center, School of Physics, Nankai University, Tianjin* 300072, *China*

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The polymethyl methacrylate (PMMA) film doped with an azo dye ethyl-red (ER) film is employed to demonstrate the properties of an all-optical switch by its photoinduced dichroism and birefringence. We show how to enhance remarkably the modulation depth of all-optical switches almost to 100% by using two linear polarization beams: one beam is inclined at 45° with respect to the probing beam and serves as a pumping beam, and the other beam is perpendicular to the probing beam and used as an erasing beam. Furthermore, a maximum-to-minimum output intensity ratio of 2000:1 is achieved in experiment, which is very useful and important for optical storages and image displays.

Keywords: all-optical switch, azo dye ethyl-red film, modulation depth

PACC: 4280V, 4225L

1. Introduction

Photochromic organic polymers have received much attention these days due to their potential applications in optoelectronic devices, optical storages, all-optical modulators, routers, time-delay relays, time-dependent switches, new filters, etc.[1−13] Azo dye is one of the most promising candidates of photochromic organic polymers for applications in all-optical switching of reversible optoelectronic devices because of its advantages, such as large electro-optic coefficients, low cost, flexibility in fabrication, fast response, higher sensitivity and easy change of the structure of its molecules.[14−19] Though various logic gates and switches were achieved by using the nonlinear photoinduced anisotropy of photochromic organic polymers were reported,[20−22] most of the previous work showed that the contrast ratio of the all-optical switch was in a range of 80:1∼300:1, which is inadequate for practical applications.[23] The modulation depth also plays an important role in improving the characteristics of the switch, and a modulation depth of 72% was reached through selecting solvent and proper concentration.[24] In the present study, two linear polarization pumping beams are used alternately to enhance the modulation depth of the all-optical switch almost by 100%, and a contrast ratio of about 2000:1 is achieved which is the highest contrast ratio for photochromic organic polymers, to our knowledge. The all-optical switch demonstrated in the paper is also useful for data storages and optical displays.

2. Experiment setup and principle

The experiment setup is shown in Fig. 1. A He–Ne laser beam with 632.8 nm was used as a probing beam, which passed through cross polarizers P1 and P2 and arrived at a detector. The sample was placed between the two polarizers P1 and P2. The wavelength of probing beam, 632.8 nm, is far from the absorption peak, in order to avoid photoinduced dichroism and to achieve a large contrast ratio, which is maximum-to-minimum output intensity ratio. The

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†Corresponding author. E-mail: Julie_chen6323@yahoo.com.cn
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polarization direction of the polarizer P3 was inclined at an angle of 45° with respect to the polarized direction of P1. A semiconductor laser (532 nm, CW) was used as a source of pumping beams 1 and 2. In order to make the pumping beams 1 and 2 illuminate alternately the sample, a special chopper was used. A metal layer was coated on the blades of the special chopper by a sputtering method and so the blades acted as mirrors to reflect a laser beam. Pumping beam 1 was from the beam (532 nm) that passed through the slots of the special chopper and reflected by a mirror and then illuminated the sample. Another pumping beam was from the beam 532 nm that hit the blades of the special chopper and was reflected by the blades. In the pathway, when the pumping beam passed through a half-wave plate, it served as pumping beam 2 and then illuminated the sample. In our experiment, the probing beam, pumping beams 1 and 2 illuminated the same area of the sample.

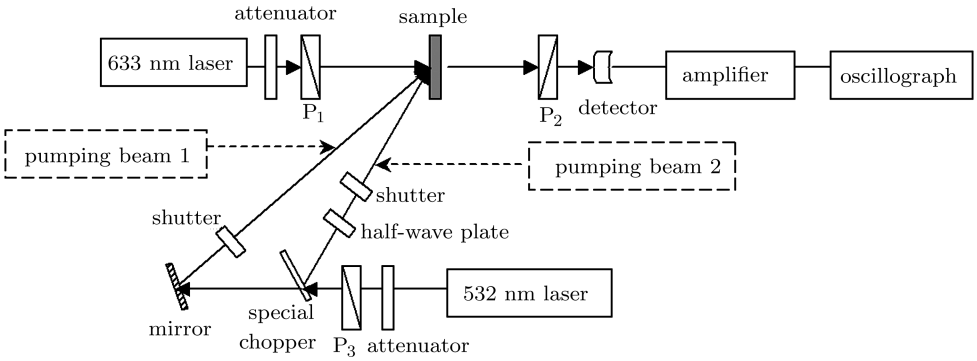


Fig. 1. Experiment setup.

The sample of azo dye ethyl-red (ER) film with high optical quality was prepared in our laboratory by using the spin-coating technique and cyclohexanone solvent. The sample was 50-μm thick and the weight ratio between ER dye and polymethyl methacrylate (PMMA) was 6 wt%. Figures 2(a) and 2(b) show the molecular structure and the absorption spectrum of ER, respectively. The absorbance peaks locate at 300 nm and 500 nm separately. On absorption of light energy around 500 nm, the ER molecules could be pumped from the trans (ground) state to cis (exciting) state, then photoisomerized. The isomerize process could be simply considered as a two-energy-level system and be expressed as

$$\text{trans} \underset{h\nu,\ \text{thermal}}{\overset{h\nu}{\rightleftharpoons}} \text{cis}\ ,$$

where the isomerization of trans to cis happened when irradiated by the 500-nm wavelength beam, and then the isomerization of cis to trans happened by further absorbing photons or thermally relaxing to the initial state[24−26] because the cis was not a stable state. If we simplify the physical processes to two energy levels: one was without the pumping beam illuminating the ER molecules, which were in ground state B, and the other was with the pumping beam illuminating the ER molecules, which were in excited level M.

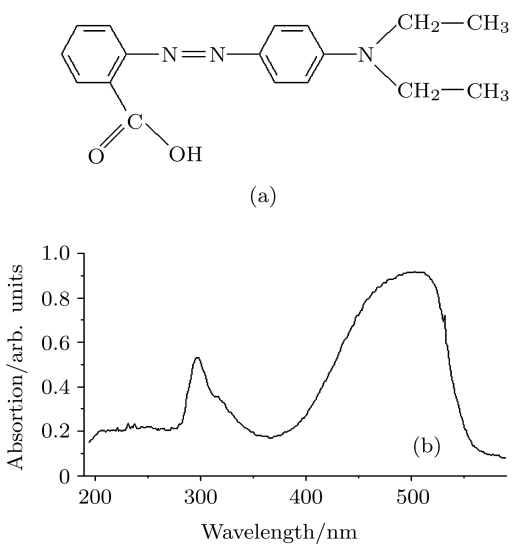


Fig. 2. Molecular structure of ethyl red (a) and absorption spectrum of PMMA doped with ethyl red at a dye concentration of 6 wt% (b).

While the ER film was illuminated by the polarized 532-nm wavelength beam, only these ER molecules with their axis orientation parallel to the electric vector of the illuminating beam absorb light and then were isomerized from trans form to cis form, which induced a change of the dimensional structure of ER molecules. The ER molecules in the unstable cis state thermally went back to trans or absorbed photons then went back to trans, and the dipole moments of ER molecules were changed since the ER molecules in cis form rotated more easily than in trans form. While the dipole moments became perpendicular to the polarization direction of the pumping light, the ER molecules did not interact with the pumping light field.[27] Thus, after the sufficient irradiation of ER chromophores by 532-nm beam, the repeated process of trans-cis-trans resulted in dipole moments of ER molecules in the direction perpendicular to the polarized direction of the pumping beam (532 nm). Finally, transition dipole moments of ER molecules perpendicular to the polarization direction of the pumping light were enhanced. Thus, it was this local dipole moments alignment that induced the ER molecules with photoinduced anisotropy in the medium.

Without the pumping light, the linearly polarized probe light illuminated the sample and cannot pass through P2, so the output light power is zero. We understand that dipole moments of ER molecules in ground (B) state were in random distribution and the medium was isotropy, therefore, no probing light passed through the P2 polarizer. The polarized pump beam 1 whose wavelength 532 nm was close to the absorption peak (500 nm) illuminated the sample, due to ordering effect of the ER dipole moments, photoinduced dichroism and birefringence occurred in the sample. Ownng to the wavelength of the probing light beam 632.8 nm was far from the absorption peak of ER molecules, the photoinduced dichroism was small. When the probing light in the polarized direction parallel to P1 passed through the ER film, the transmitted probing beam with the ellipse polarization due to the photoinduced birefringence was affected in the ER film; thus, the transmitted probing beam with the polarized component perpendicular to P1 passed through P2, and the output signals could be achieved. The output depended not only on the intensities of the probing beam and pumping beam 1 but also on their polarized directions. The highest maximum-to-minimum output intensity ratio could be obtained by the mechanism of optimization-direction-selected photoisomerization of ER dipole molecules controlled by the polarized beams and their intensities.[22]

When the ER molecules were illuminated by pumping beam 1, a part of ER dipole molecules were pumped from B state to M state through the photoisomerization process, and the directions of the dipole moments would be redistributed to be along the direction perpendicular to the polarized direction of pumping beam 1, correspondingly, and the refractive index and the absorption coefficient of ER molecules would be changed as well. The change of refractive index would modulate the photoinduced birefringence, and the change of absorption coefficient would modulate the photoinduced dichroism. Considering the characteristics of photoinduced birefringence and dichroism, the dynamic transmitted output intensity can be described as follows:[22]

$$\begin{aligned} & I_{\rm out}(t) \\ = \; & I_{\rm in}10^{-\Delta D}\sin^2(2\beta_i)\sin^2(\pi\Delta n d/\lambda_{\rm probe}) \\ & \times \exp^{-2.3026[\varepsilon^{\parallel}_{\rm B-probe}N_{\rm B}(t)+\varepsilon^{\parallel}_{\rm M-probe}N_{\rm M}(t)]d}, \end{aligned} \quad (1)$$

where $I_{\rm in}$ is the incident probing beam intensity, $\lambda_{\rm probe} = 632.8$ nm is the probing beam wavelength, $\Delta D = 1.15$ represents the effective photoinduced dichroism and Δn denotes the effective photoinduced birefringence. In the calculation, $\Delta n = A(\partial N_{\rm M}(t)/\partial I_{\rm p})\Delta I_{\rm p}$, where $\Delta I_{\rm p}$ is the intensity change of the pumping beam passing through the ER film, A is a constant and $d = 50$ μm is the film thickness. $\varepsilon^{\parallel}_{i-{\rm probe}}$, i =B (or M) represents the molecular extinction coefficient induced by absorption cross section in the polarization direction of probe beam at probing wavelength in B (or M) state. $N_{\rm B}(t)$ and $N_{\rm M}(t)$ are the molecular concentrations of the B state and the M state respectively. $N_{\rm B}(t) + N_{\rm M}(t) = N(t)$, where $N(t)$ is the total molecular concentration; at angle α (see Fig. 3), $N_{\rm B}(t) + N_{\rm M}(t) = N(t)/2\pi$. Without the pumping light, $\Delta n \approx 0$, there were no output signals. Only when ER was illuminated by the pumping beam 1, there were output signals. β_i, i= 1, 2 was the angle between the probing beam and pump beams 1 and 2, respectively, and when $\beta_1 = 45°$ (see Fig. 3), the $I_{\rm out}$ reached its maximum. According to the population rate equation for simplified two-energy levels,

$$N_{\rm M} = (Nk_1/2\pi k)[1 - \exp(-kt)], \quad (2)$$

where parameter $k = k_1 + k_2 + k_3$, with $k_3 = 1/\tau$, τ being the lifetime of the M state, k_1 and k_2 being the effective different photochemical rate constants, which

can be written as a function of the film optical properties and the intensities of the illumination beams, could be calculated to be

$$k_1 = 2.3026\phi(\varepsilon_{\mathrm{B}1}\lambda_1 I_1 + \varepsilon_{\mathrm{B}2}\lambda_2 I_2)/N_{\mathrm{a}}^h c, \quad (3)$$

$$k_2 = 2.3026\phi(\varepsilon_{\mathrm{M}2}\lambda_2 I_2 + \varepsilon_{\mathrm{M}1}\lambda_1 I_1)/N_{\mathrm{a}} hc, \quad (4)$$

where $\phi = 0.45$ is the quantum yield of the photo transformation, $\lambda_1 = 632.8$ nm, $\lambda_2 = 532$ nm, N_{a} is the Avogadro's number, h is the Planck's constant, c is the speed of light (ms^{-1}), $\varepsilon_{\mathrm{B}1}$ and $\varepsilon_{\mathrm{B}2}$ are the effective molecular extinction coefficients of ER in B state for B→M and can be estimated by $\varepsilon = -\log(T)/dN$ with d and T being the thickness and the transmittance of the sample, N is the molecular concentration, $\varepsilon_{\mathrm{B}}^{533} = \varepsilon_{\mathrm{B}2} = 3.5 \times 10^6\ \mathrm{L\cdot mol^{-1}\cdot cm^{-1}}$, $\varepsilon_{\mathrm{B}}^{633} = \varepsilon_{\mathrm{B}1} = 3000\ \mathrm{L\cdot mol^{-1}\cdot cm^{-1}}$, $\varepsilon_{\mathrm{M}}^{533} = \varepsilon_{\mathrm{M}2} = 3.4 \times 10^6\ \mathrm{L\cdot mol^{-1}\cdot cm^{-1}}$, $\varepsilon_{\mathrm{M}}^{632.8} = \varepsilon_{\mathrm{M}1} = 0$ are the effective molecular extinction coefficients of ER in B state for M→B, I_1 and I_2 ($\mathrm{mW/cm^2}$) are the intensities at wavelengths λ_1 and λ_2, respectively. In calculation, we assume

$$\varepsilon_{\mathrm{B}i} = 10\varepsilon_{\mathrm{B}i}^{\parallel}, \quad (5)$$

$$\varepsilon_{\mathrm{M}i} = 10\varepsilon_{\mathrm{M}i}^{\parallel}. \quad (6)$$

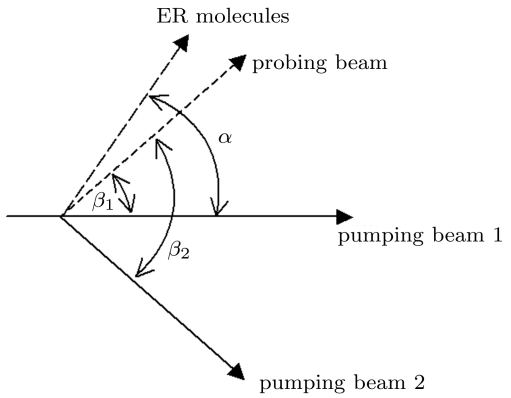


Fig. 3. Angle configuration of the ordering direction of ER molecules and the polarization directions of pumping and probing beams.

In our experiment, in order to enhance the modulation depth, the output should be decreased very sharply when pumping beam 2 is turned on and pumping beam 1 is turned off simultaneously. Note that the polarized direction of pumping beam 2 is perpendicular to that of probing beam 1. It is easy to understand that when the ER molecules are irradiated by polarized pumping beam 1 and the transition dipole moments of photoisomerization ER molecules finally are perpendicular to the polarized direction of pumping beam 1, pumping beam 1 does not interact with the orientated ER molecules. However, the direction of the polarized probing beam was not perpendicular to the dipole moments, and there were interactions between the polarized probe light and the orientated ER molecules induced by pumping beam 1 to generate photoinduced anisotropy. When pumping beam 1 was turned off and pumping beam 2 was turned on simultaneously, the orientated ER molecules induced by the pumping beam 1 would re-select the direction under the condition of pumping beam 2. Sufficient processes of trans-cis-trans resulted in the dipole moments of ER molecules in the direction perpendicular to the polarized direction of pumping beam 2. Note that the dipole moments of ER molecules were parallel to the polarized direction of the probing beam, and the ordered dipole moments of ER molecules made no contribution to the transmitted polarization direction of P2, thereby, the output signals were zero. In the absence of pump beam 1 and the sample was illuminated only by pumping beam 2 at an angle of 90° (between the probing beam and pumping beam 2, see Fig. 3), that is, in Eq. (1), $\sin^2 2\beta_2 = 0$, thus, the I_{out} would finally be zero, namely, $I_{\mathrm{out}} = 0$. On the basis of this principle, we designed our experiment as follows. When pumping beam 1 was turned off and pumping beam 2 which was in the polarization direction perpendicular to that of probing beam, was turned on, most of the dipole moments for ER molecules would be perpendicular to the polarization direction of pumping beam 2 as long as the intensity of pumping beam 2 was strong enough, and the output signals would quickly weaken.

The scheme of the polarization direction of ER dipole moments is shown in Fig. 3. As described above, when pumping beam 1 was turned off and pumping beam 2 was turned on, the output would be close to zero in a certain time, which depended on the intensity of pumping beam 2. The stronger the intensity of pumping beam 2 was, the weaker the intensity of output signal would be. The output signal quickly decayed to a very low value when pumping beam 2 was strong enough. By this method, high modulation-depth-switch could be easily obtained.

3. Experimental results

In our experiment, we found that for a fixed probing beam, when the pumping beam intensity was low, the transmitted power of the probing beam was proportional to the pumping beam intensity. While the

pumping beam intensity was about 100 mW/cm^2, the output saturation was reached. Similarly, for a fixed pumping light, when the probing beam intensity was low (high), the output was low (high) because the transmission was low (high). When the probing beam intensity was strong enough, the output saturation was reached, which indicated that when the pumping beam increased, the output did not change for a fixed probing beam. The highest maximum-to-minimum output intensity ratio of 2000:1 was achieved when the optimal intensity of the probing beam was 40 mW/cm^2 and that of the pump beam 1 was 400 mW/cm^2, and the corresponding experimental results are shown in Fig. 4. Here, the signal light intensity can reach a maximum value when the angle included between the polarization directions of the probing beam and the pumping beam 1 is 45°, an optimal angle. With only the probing beam and pumping beam 1, the dynamic output signals are shown as curve 1 in Fig. 4, which is background data without pumping beam 2. Note that curve 1 decays slowly. It is easy to understand that the slowly decaying curve will reduce the modulation depth of an optical switch. After pumping beam 1 was turned off, pumping beam 2 was turned on with intensities of 40, 160, 240, 320 and 400 mW/cm^2, whose corresponding output curves are shown as curves 2, 3, 4, 5 and 6, respectively. Note that the stronger the intensity of pumping beam 2 is, the faster the output will decay. When pumping beam 1 was turned off, a substantial number of ER molecules remained "frozen" in their orientations, which would take some time to thermally go back to their initial states. Therefore, the output signals decayed slowly. After pumping beam 1 was turned off and only pumping beam 2 illuminated on the sample the dipole moments of the oriented ER molecules by pumping beam 1 would rotate quickly to the directions perpendicular to the polarization direction of pumping beam 2, thus, the oriented or re-selected directions of the dipole moments of ER molecules were parallel to the direction of the probing beam, and the intensity of the transmitted probing beam decreased quickly to zero. This effect of pumping beam 2 was similar to an optical erasing. This resulted in the faster decay of outputs since the optical "erasing" process was much faster than the thermally relaxing process of ER molecules. While the weaker probing beam only oriented a small part of the ER molecules whose contribution to the decay rate was small, those ER molecules oriented by pumping beam 2 dominated the decay process of the transmitted probe beam intensity.

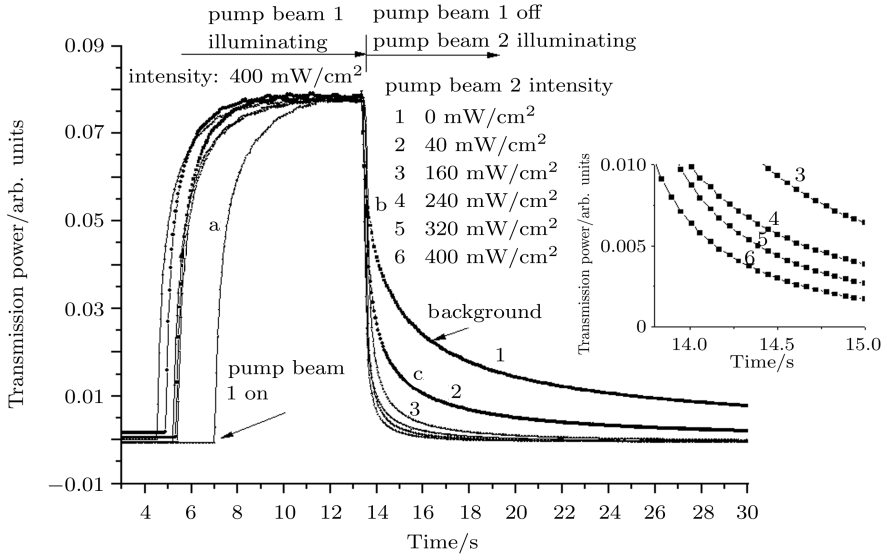


Fig. 4. Dynamic transmitted probe-beam intensities varying with time. The intensities of probing beam and pumping beam 1 are 40 and 400 mW/cm^2, respectively. Stage a: the exciting process of pumping beam 1. Stage b: the "erasing" process of pumping beam 2 for curves 2, 3, 4, 5 and 6. Curve 1 shows the decay process without pumping beam 2. Curves 3, 4, 5 and 6 in the inset show the decay stages, their corresponding intensities of pumping beam 2 are 160, 240, 320 and 400 mW/cm^2.

Curve 1 in Fig. 4 shows that the highest modulation depth is 87.5% at the cost of longer time (∼14 s). Adding pumping beam 2 with an intensity of 40 mW/cm^2, based on curve 2, the modulation depth reached 87.5% within 2 s, and the highest modulation depth reached 96%. Similarly, for pumping beam 2 with intensities of 160, 240, 320 and 400 mW/cm^2, the highest modulation depths were over 96%. Especially, when the intensity of pumping beam 2 was 400 mW/cm^2, the modulation depth reached almost 100%. The modulation depths corresponding to Fig. 4 and their required times are shown in Table 1. The required time shows a remarkable dependence on the intensity of pumping beam 2. We can see that the required time for a 75%-modulation depth can be shortened from 4.15 to 0.213 s by pumping beam 2 with an intensity of 400 mW/cm^2. The modulation depth could finally reach 100% when the intensity of pumping beam 2 reached 400 mW/cm^2.

The dynamic output signals first increased from a very low value to a stable value. Then after pumping beam 1 was turned off and pumping beam 2 was turned on, the output signals started to decay. Without pumping beam 2 and when the probing beam with an intensity of 40 mW/cm^2 and pumping beam 1 with an intensity of 400 mW/cm^2 illuminate on the ER film, the output results are shown as the open circles in Fig. 5, which comes from curve 2 in Fig. 4. The theoretical calculation results are shown as a solid line in Fig. 5, and the results of the calculation and experiment are accordant with each other. For different intensities of pumping beam 2, the decay stages of the output intensities can be expressed as

$$A(t) = A_1 \mathrm{e}^{-K_1 t} + A_2 \mathrm{e}^{-K_2 t} + A_{\mathrm{th}}, \qquad (7)$$

where $A(t)$ is the optical intensity in a decay stage, A_1 and A_2 are the contributions to the initial optical intensity, K_1 and K_2 are the rates of fast and slow components, respectively, and A_{th} is the remnant birefringence when time approaches infinity. This model has been widely used to fit photochromic decoloration.[28−30] The decay segments obtained at each power level fit well to the exponential functions. The decay rate depends on K_1 and K_2, which is obtained by the least square fitting using the experimental data. From the fitting of experimental data, the deduced results of the constant values of K_1 and K_2 corresponding to several powers of pumping beam 2 are shown in Fig. 6. It is seen that the rate constant K_1 increased faster than K_2 when the power of pumping beam 2 was increased. K_1 and K_2 were functions of I_2 (pumping beam 2) and expressed as

$$K_1 = 0.00695 + 0.0000306 I_2, \qquad (8)$$

$$K_2 = 0.00084 + 0.00000343 I_2. \qquad (9)$$

The stronger the intensity of pumping beam 2 was, the faster the decay time and the larger the modulation depth of all-optical switch would be.

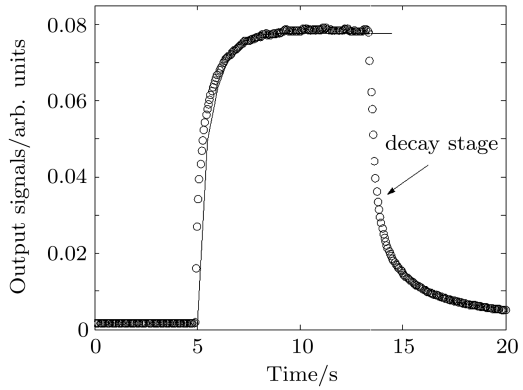


Fig. 5. Comparison between results of experimental measurement (open circles) and theoretical calculation (solid curve).

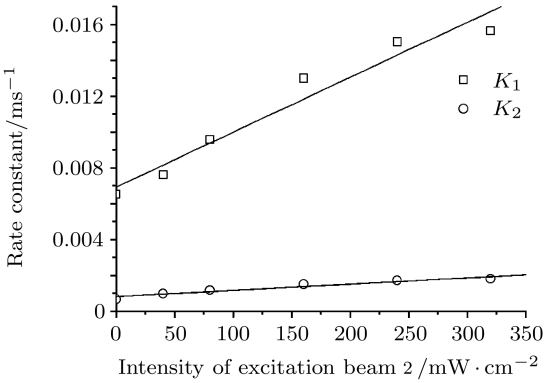


Fig. 6. Rate constants versus the intensity of pumping beam 2, with the intensity of pumping beam 1 fixed at 400 mW/cm^2.

Table 1. Decay time as a function of intensity of pumping beam 2.

pump beam 2/mW · cm^{-2}	50%/sec.	75%/sec.
0	0.758	4.155
40	0.274	1.085
160	0.218	0.440
240	0.157	0.265
320	0.117	0.226
400	0.107	0.213

Figure 7 shows the characteristics of the demonstrated all-optical switch. The intensities of the probing beam and pumping beam 1 were 40 mW/cm^2 and

400 mW/cm^2, respectively. The first row (Fig. 7(a)) shows the output signals only under the condition of pump beam 1, indicating that the decay rate was slow, and the modulation depth could only reach ∼52% within 0.72 s. The modulation depth and its required time depended on the decay rate of the output signals, which could be changed by pumping beam 2 with an appropriate intensity. After pumping beam 1 was turned off and pumping beam 2 with an intensity 240 mW/cm^2 was turned on, the output signals as a function of time turn into those shown in the second row (Fig. 7(b)). In this case, the modulation depth could reach 86.5% within 0.1 s. The pulse train of pumping beam 1 modulated by the special chopper is shown in the third row (Fig. 7(c)). It is clear that the modulation depth can be enhanced greatly in a short time by using our scheme.

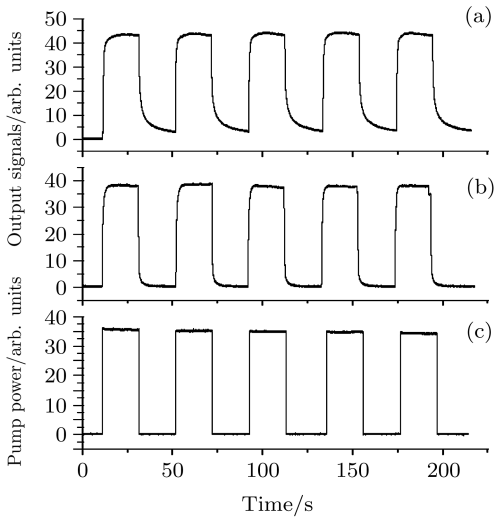


Fig. 7. Characteristics of the demonstrated all-optical switches.

4. Summary

With an azo dye-ethyl red film, we can enhance the modulation depth of an all-optical switch by linearly pumping beams 1 and 2. A very fast decay rate is obtained due to the fact that the optical excitation is more efficient in changing the direction of the molecular dipole moment than the thermal effect, which can greatly enhance the response time of the all-optical switch. As a result, a very high contrast ratio (about 2000:1) is achieved, which is useful for optical storages and displays. In addition, if we control the thickness of the ER film so as to satisfy the relation $\pi\Delta nd/\lambda_{\rm probe} = \pi/2$, according to Eq. (1), we can obtain an even higher contrast ratio. The method we reported here is easy to use, and it is useful for improving the characteristics of all-optical switches, by which high contrast and high modulation depth of output signals are both obtained at the same time. Although the switch we have demonstrated is not a "fast" enough switch, this does not affect the improvement of the characteristics of an all-optical switch. In addition, a faster-response material can be obtained by intrinsic material modification,[31−37] which can solve the slow response-time problem.

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