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MOLECULAR PHOTONICS BY FEMTOSECOND LASER

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Photochromic spiropyran in guided wave mode geometry was used to demonstrate the performance of an ultrafast molecular photonics system utilizing femtosecond (fs) white light as a probe and fs blue light as a pump. Specific and reversible changes were observed by photochromism in fs white light for dip wavelength and/or dip reflectance depending on the incident angle.The present system will contribute a great deal to future ultrafast all-optical processing in a broad wavelength region.

INTRODUCTION

Novel materials, devices and systems are required for much faster data processing speed and much higher recording density due to rapidly expanding volume of information and data. Molecules and photons have many superior properties for all-optical parallel data processing as schematically shown in Figure 1. Through interactions of molecules or molecular assemblies with photons, such properties can be directly converted to changes in physical properties of materials such as fluorescence, absorption, refractive index, conductivity, or optical nonlinearity. Absorption changes due to excited state formation, photochromism, or photoinduced electron transfer are some examples among them. We have been making efforts to develop new molecular photonics materials and devices by optically controlling electronic states of various organized molecular systems and by making use of photoinduced complex refractive index changes for optical parallel processing [1–12]. We have demonstrated the switching ON in a nanonsecond (ns) laser pulse width and the switching OFF in a

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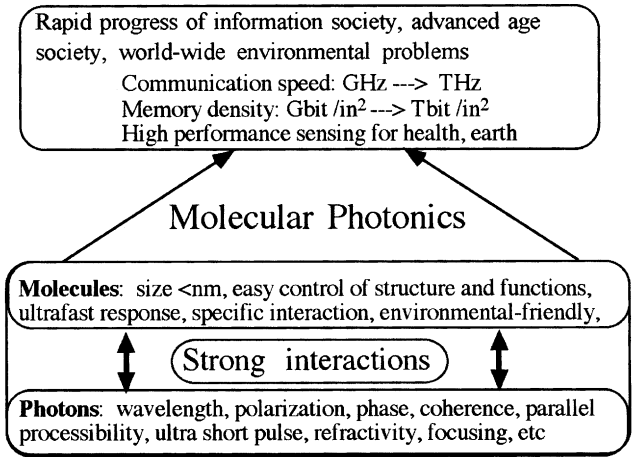

FIGURE 1 In order to fully utilize superior properties of molecules and photons.

very wide time range from 30 ns to infinity depending on the nature of photoresponsive materials[1–6]. In the present paper, some of our recent achievements on materials and devices for all-optical ultrafast molecular photonics will be reported.

Our all-optical spatial light modulator (SLM) based on complex refractive index changes upon photoexcitation of an organic dye-doped polymer thin film can make fs response in principle, because we use resonance condition changes of the guided optical waves (guided wave mode) in the ATR geometry based on photoinduced changes of an imaginary and/or a real part of the refractive index. It is also possible to expand the wavelength to near infra-red region compatible with the present light communication system by developing materials showing fast and large absorption changes in 1300–1700 nm region. We have been making two approaches to achieve such properties by temporally expanding the electronic delocalization upon photoinduced electron transfer or by making charge resonance interactions [7–11]. We have achieved wavelength responses upto about 2600 nm [9,10] and time responses with less than 1 picosecond (ps) for switching ON (forward electron transfer) and about 3 ps for OFF (reverse electron transfer) [8].

EXPERIMENTAL

A rotating stage with a prism and a sample glass slide was set in a femtosecond (fs) pump-probe transient absorption measurement system as schematically shown in Figure 2. The second harmonics (400 nm, 10 Hz) of

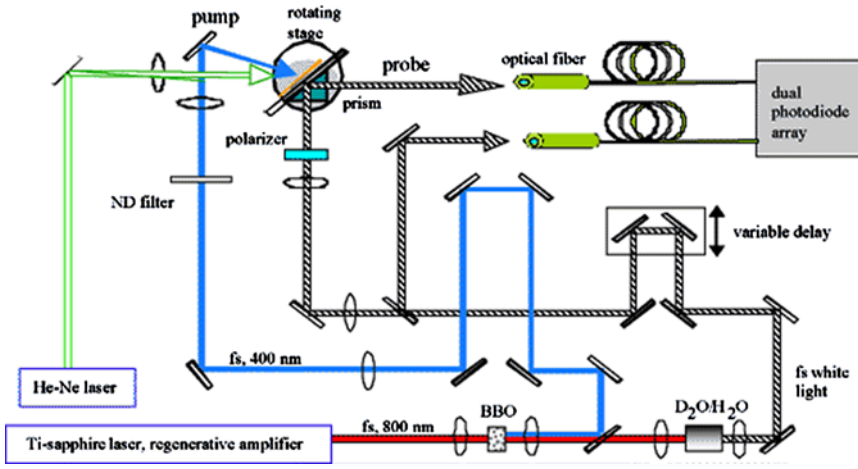

FIGURE 2 Schematic representation of the fs pump-probe reflection measurement system. (See COLOR PLATE I)

amplified fs Ti:sapphire laser was used to excite spiropyran (SP) in Arton (JSR Co. Ltd.) film spin-coated on a silver thin film (50 nm). Upon excitation at 400 nm, SP photoisomerized to photomerocyanine (PM), which was stable and was reversibly photoisomerized again by irradiation with green He–Ne laser. The fs probe white light obtained by focusing the residual 800 nm light into a cell containing D_2O/H_2O (2:1) mixture after passing through a BBO crystal to generate the second harmonics was p-polarized by a fs polarizer and was introduced from a prism side after an optical delay system. The reflected white light and the incident one were detected by a Photonic Multi-channel Analyzer (Hamamatsu Photonics) system with a dual photodiode array.

RESULTS AND DISCUSSION

A profile of fs white light is shown in the inset (a) of Figure 3. A dip was found in it at a given wavelength as shown in the inset (b) of Figure 3 as an example for a 360 nm thick film of SP/Arton (1:2) at the incident angle of 53.0°, which was attributed to the guided wave mode. The observed dip was broader than those monitored by CW lasers because of the extremely short pulse width (ca. 160 fs). The dip wavelength shifted to a longer side by decreasing the incident angle from about 500 nm at 65.0° to about 760 nm at 39.0° as shown in Figure 3. The average dip shift was 10 nm/degree, so we can easily tune the wavelength at which we want to make all-optical modulation with our guided wave mode device.

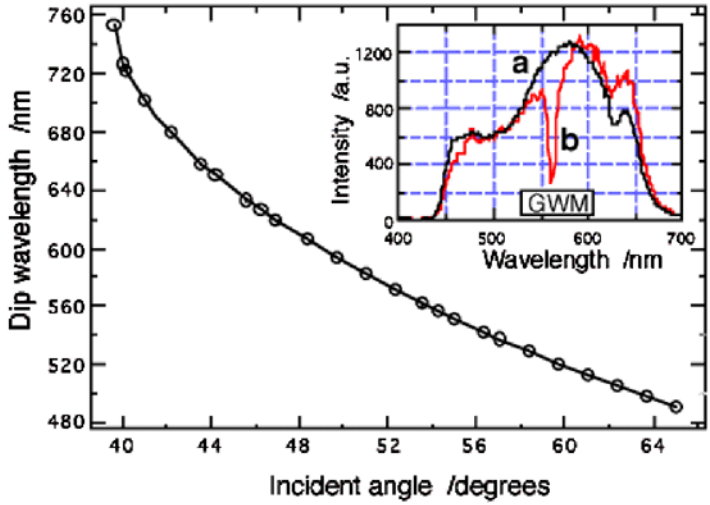

FIGURE 3 Relationship between the dip wavelength and the incident angle for a spin-coated 360 nm thick film of SP/Arton (1:2) observed by fs white light. The inset shows profiles of fs white light without (a) and with (b) a spin-coated film at the incident angle of 53.0 degrees. (See COLOR PLATE II)

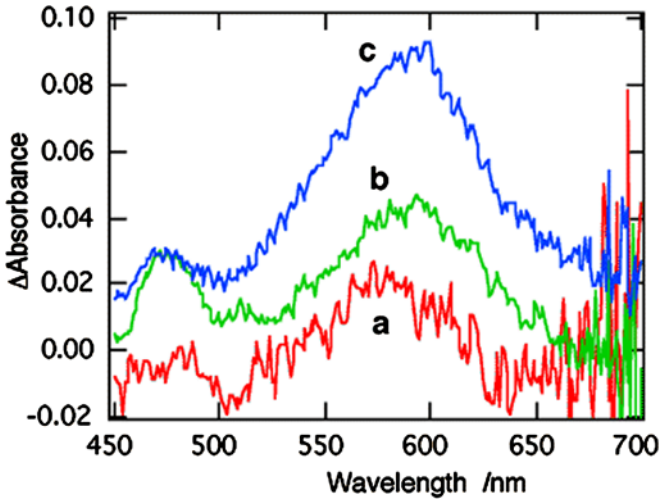

FIGURE 4 Difference absorption spectra of 360 nm thick film of SP/Arton (1:2) upon fs laser irradiation at 400 nm for (a) 2, (b) 10, and (c) 16 s. (See COLOR PLATE III)

This film showed highly efficient photochromism upon fs laser excitation at 400 nm as shown in Figure 4 for (a) 2, (b) 10, (c) 16 s irradiation at 10 Hz repetition. The dip wavelength and/or minimum reflectance showed specific changes by photochromism upon excitation with fs laser at 400 nm and returned to the original value by reverse photochromism upon He–Ne laser irradiation as shown in Figure 5 by arrows. Typical changes of reflected white light spectra are shown in Figure 5 at four incident angles; (a) 53.0, (b): 50.0, (c): 42.6, and (d) 39.6°, respectively. The irradiation time at 400 nm was (a) 2–10 s, (b) 1–10 s, (c) and (d) 3–16 s, respectively. The "direction" of changes of reflectance clearly depended on the incident angle or the dip wavelength. By photochromism from SP to PM, the reflectance increased with shifting the dip wavelength to a shorter side at 53.0° and to a longer side at 42.6°, or without shifting at 50.0°. At the incident angle of 39.6°, the dip wavelength shifted to a longer side with almost no changes of reflectance. Such changes corresponded well with complex refractive index changes by photochromism between SP and PM forms as shown in Figure 6. All reflectance changes were highly reversible for many times.

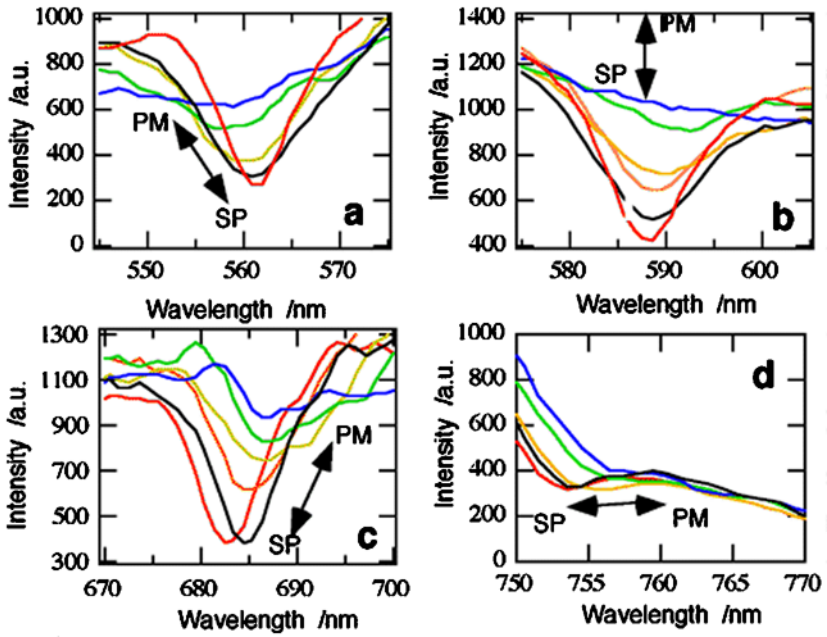

FIGURE 5 Changes of reflected light intensity for SP/Arton (1:2) film (360 nm thick) upon fs laser excitation at 400 nm and subsequent He–Ne laser irradiation at 543.5 nm. The incident angle is (a) 53.0, (b) 50.0, (c) 42.6, and (d) 39.6°, respectively. (See COLOR PLATE IV)

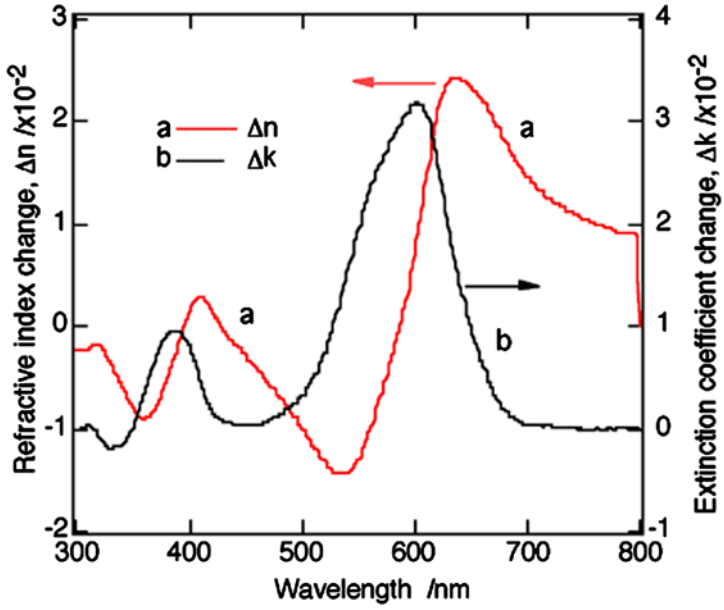

FIGURE 6 Wavelength dependence of (a) refractive index and (b) extinction coefficient of a polymer thin film containing SP upon photochromism. (See COLOR PLATE V)

CONCLUSION

A new ultrafast all-optical parallel processing system was developed by the use of fs pump and probe arrangement, Spiropyran was used to demonstrate the performance of this system utilizing fs white light as a probe for guided wave mode. The present system will contribute a great deal to THz all-optical parallel processing over a wide wavelength region covering near infra-red region by appropriate combination of transient photoresponsive materials we have developed.

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