Extended Abstracts of the 2002 International Conference on Solid State Devices and Materials, Nagoya, 2002, pp. 220-221

C-5-1 (Invited)

Ultrafast Molecular Photonics for All-Optical Data Processing

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1. Introduction

Due to rapidly expanding volume of information and data, novel materials, devices and systems are required for much faster data processing speed and much higher recording density. Most prominent way to achieve such a goal will be optical parallel data processing such as photon-mode optical memory or optical correlation. A spatial light modulator (SLM) is one of the key devices in optical parallel processing. Presently available SLMs are based on liquid crystals (LC), so the response is not so fast, typically millisecond (ms) - sub ms. Several "all-optical" photo-addressed SLM were reported. The response times of these devices, however, were even slower than LC-SLM due to a thermal process and many efforts are to be made to improve them for for on-line optical correlator or dynamic holography.

Through interactions of molecules or molecular assemblies with photons, many superior properties of photons 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 making various organized molecular systems and by optically controlling their electronic states. In the present article, some of our recent achievements will be discussed aiming at all-optical ultrafast molecular photonics.

2. Ultrafast Photoresponsive Materials

We have achieved novel photoinduced electrochromism which is color changes due only to the photoinduced electron transfer and reverse reactions [1,2]. Molecular control of the lifetime of colored species over extremely wide range from 1 ps to infinity was demonstrated by ion-pair charge transfer complexes of 4,4'-bipyridinium ions with various counter anions. A very wide range of the wavelength of colored species was also achieved in the visible and near-infrared region upto about 2600 nm. Photon-mode super-resolution to exceed the diffraction limit of light in optical memory was also achieved based on transitory photobleaching of phthalocyanine derivatives [1,2].

3. All-Optical Parallel Data Processing

In order to fully utilize such photoinduced changes of optical properties in molecules, we have proposed a novel all-optical device which can be used as a photon-mode SLM or memory based on complex refractive index changes upon photoexcitation of an organic dye-doped polymer thin film as schematically shown in Fig. 1 [3].

In principle fs response can be achieved in this system, because we use resonance condition changes of the guided optical waves (guided mode) in the ATR geometry based on the changes in an imaginary or a real part of the refractive index as shown in Fig. 2 (from **a** to **c** or **a** to **b**) due to transient absorption or its Kramers-Kronig transformation. The main advantages using the guided mode are (1) its high sensitivity to small changes in refractive index and thickness, and (2) its sensitivity to both p- and s-polarized light. So far we have achieved repeated

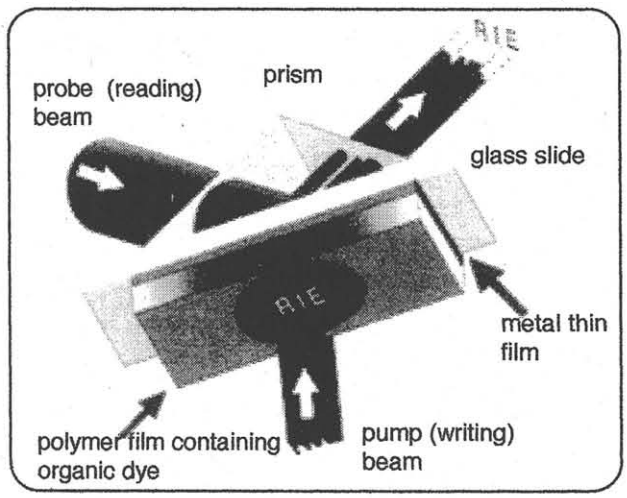

Fig. 1 Schematic representation of all-optical device based on photoinduced complex refractive index changes in guided mode geometry.

light modulation using pulsed nanosecond (ns) laser and CuPcS and ZnPcS in guided wave-mode geometry. The response time was controlled by the triplet lifetime of phthalocyanines, 30 ns for CuPcS and 0.55 ms for ZnPcS [4]. We are making efforts to achieve much faster responses using fs laser and appropriate materials.

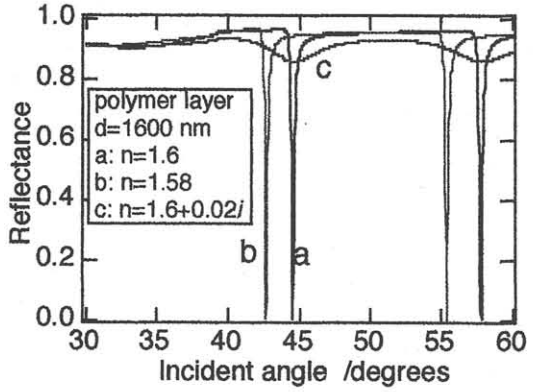

Fig. 2 Calculated incident angle dependence of reflectance for a polymer film on a silver film (50 nm) upon changes of complex refractive index.

We have also demonstrated ultrafast parallel optical recording based on the same geometry and using photochromic compounds instead of phthalocyanines. Strong absorption in the visible region due to photomerocyanine (PM) formed by UV irradiation can be held for a long time and be reverted to that of spiropyran derivative (SP) by visible irradiation. Spectra of extinction-coefficient and refractive-index changes (Δk and Δn) of polystyrene thin film containing SP upon UV excitation were evaluated from the observed difference absorption spectra before and after UV, and its Kramers-Kronig transformation. The extinction-coefficient and/or refractive-index changes over a wide wavelength range approximately from 400 to 800 nm can be utilized to operate a wide range all-optical switch. We have achieved parallel recording of an image by a single shot ns laser and also the phase shift. In addition to very fast photoresponses, it is essential to write and read a two-dimensional image pattern for the optical parallel data processing. Fig. 3 shows the microscopic photographs (x 500) of the image written through a USAF Test Target as a mask by a single shot of ns 355 nm laser (1 mJ/pulse) on a spiropyran-doped PS film deposited on a silver film. At least 128 line pairs/mm was clearly seen, which corresponds to the spatial resolution better than 3.9 μm.

4. Conclusion

The guided mode geometry fully utilizing photoinduced changes of molecular electronic state will contribute a great deal for ultrafast molecular photonics and all-optical data processing in the near future.

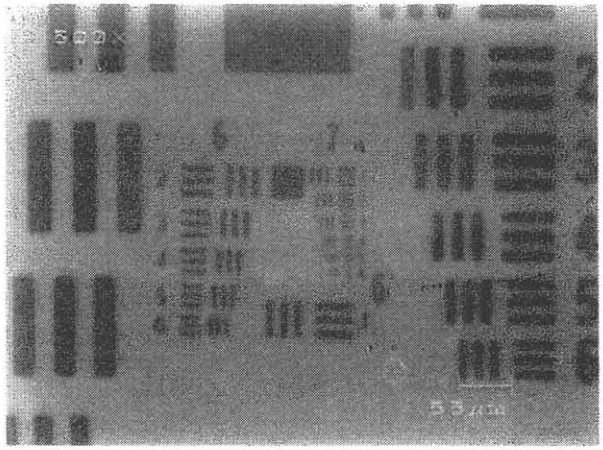

Fig. 3 Image (x 500) written through a USAF Test Target as a mask by a single shot of ns 355 nm laser (1 mJ/pulse) on a spiropyran-doped poly(styrene) film deposited on a silver film.

Acknowledgments

The author would acknowledge H. Sakaguchi, H. Kawai, T. Hamada, K. Sasaki, H. Inoue, F. Iizuka, T. Adachi, S.H. Park, I. Yoshida, K. Yamaji, A. Naito, and Prof. N. Okamoto of Shizuoka University, Prof. M. Hanack and Dr. H. Heckmann of University of Tübingen, D. Matsunaga of Nippon Kayaku Co. Ltd. for their contribution to the present work. Partial supports by the Grant-in-Aids for Scientific Research on Priority Areas "Molecular Synchronization for Design of New Materials System" (No. 11167242, 13022230) from the Ministry of Education, Science, Sports and Culture, Japan are greatly acknowledged

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