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Photoinduced electron transfer in molecular hetero- Langmuir-Blodgett (LB) films consisting of Green fluorescent protein (GFP)/viologen/TCNQ was investigated. In this work, charge transfer process in the molecular hetero-films has been investigated by femto-second transient absorption spectroscopy. Based on the transient absorption in the GFP/viologen/TCNQ series hetero-films, charge transfer from excited GFP to viologen (TCNQ) was observed with subpicosecond time resolution.

Keywords: Green fluorescent protein (GFP); Transient absorption; Langmuir-Blodgett (LB); Charge transfer

INTRODUCTION

Photoinduced electron transport processes in nature such as photoelectric conversion and long-range electron transfer in photosynthetic organisms are known to occur not only very efficiently but also unidirectionally through the functional groups of biomolecules.^[1,2]

Electron transfer in the molecular arrays is a sensitive function of donor-acceptor separation and molecular orientation. Transient absorption

measurements in the hetero-LB films have not been performed yet. In this work, charge separation process in the LB film has been investigated by femtosecond transient absorption spectroscopy. Based on the transient absorption in the S homo and S/R/A series hetero-LB films, charge separation from excited S to R (A) was investigated with subpicosecond time resolution.

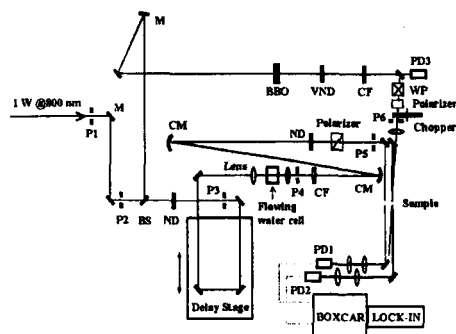


FIGURE 1. Experimental Setup for transient absorption measurement

EXPERIMENTAL DETAILS

Green fluorescence protein (GFP), viologen, and TCNQ were employed as electron sensitizer (S), relay (R), and acceptor (A) units, respectively. The deposition of LB films was carried out using a circular type Langmuir trough (Type 2022, Nima Tech., UK). The experimental setup for transient absorption measurement system was shown in Fig. 1.

A light pulse from laser system composed of a self-mode-locked femtosecond Ti:Sapphire laser (Clark MXR, NJA-5), a Ti:sapphire regenerative amplifier (Clark MXR, CPA-1000) pumped by a Q-switched Nd:YAG laser (ORC-1000), a pulse stretcher/compressor OPG-OPA

system, and an optical detection system (FWHM: 400 fs, wavelength: 400 nm, repetition rate: 20 Hz) was introduced to excite the S molecules. With fast electronics using a V/V amplifier of 300 MHz frequency (Model SR445, Stanford research) and a storage oscilloscope of 500 ps resolution (2Gs/s, model 54616B, Hewlett-Packard), the interlayer photocarrier movement was detected as voltage built up between the electrodes which form a 50 Ω strip line geometry in order to acquire signals with high time resolution.

RESULTS AND DISCUSSION

In A (TCNQ) solution, the TCNQ⁻ monomer band at 740 and 840 nm increased as the solution concentration increased. On the other hand, in A homo-LB films, TCNQ⁰ dimer ((TCNQ)₂⁰) and TCNQ⁻ dimer ((TCNQ)₂⁻) were observed at 500 and 660 nm, respectively, due to the dimer formation in A homo-LB films. Since LB films can be deposited through head (hydrophilic functional group)-head and tail (hydrophobic alkyl chain)-tail interactions, TCNQ functional groups can exist as dimeric form. In S/R(10)/A(31) hetero-LB films, there was no new absorption band except individual S, R and A peaks, indicating that neither energy level change nor chemical reaction occurred in S/R/A hetero-LB films.

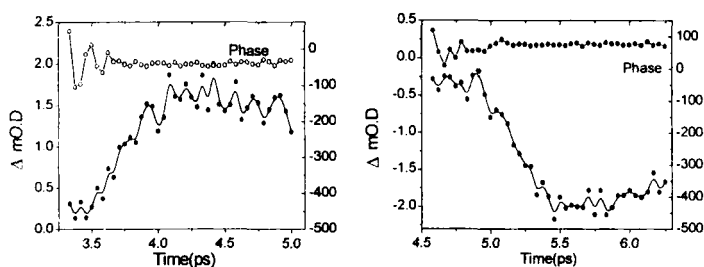


FIGURE 2. Transient absorption spectrum: a, 660nm TCNQ dimer increase, b, 500nm TCNQ⁰ dimer decrease.

Upon photoexcitation at 400 nm, the absorption change or photoinduced absorption of dimer of A ((TCNQ)₂⁰, (TCNQ)₂⁻) was observed in the transient absorption spectra(Fig.2). The transient species contributing to the transient absorption spectra were listed in Table 1. It can be confirmed that the electrons are transported from S to A via R. Since TCNQ can accept the electrons from S, the TCNQ₂⁻ bands would increase, and TCNQ₂⁰ bands would decrease. The rising time and decay time of absorption changes in TCNQ₂⁻ dimer and TCNQ₂⁰ dimer were c.a. 0.5ps.

TABLE 1. Time constants and charge transfer rates of each film

Transient signal (nm)	Suggested transient species
500	TCNQ ⁰ dimer
660	TCNQ ⁻ dimer

CONCLUSION

The fast charge separation process was clearly observed due to the better time-resolution of transient absorption technique. It is concluded that the fast electron transfer from electron sensitizer to electron acceptor can occur in sub-picosecond resolution.

Acknowledgements

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